

Neutron scattering, facilities and instrumentation

Robert Georgii

MLZ is a cooperation between:

Literature

Neutron scattering: A Primer by Roger Pynn

Los Alamos Science (1990)

<http://library.lanl.gov/cgi-bin/getfile?19-01.pdf>

Elementary Scattering Theory: For X-ray and neutron users

D.S. Sivia (2011)

St John's College, Oxford

ISBN 978-0-19-922867-6

Properties of the neutron

- Mass: $m_n = 1.675 \times 10^{-27}$ kg
- Charge = 0
- Spin = 1/2
- Magnetic moment: $m_n = -1.913 \mu_B$
- Velocity v , kinetic energy E , wavelength λ , wavevector k , moderator temperature T

$$E = \frac{1}{2}mv^2 = k_B T = \frac{\left(\frac{hk}{2\pi}\right)^2}{2m}, \text{ where } k = \frac{2\pi}{\lambda} = \frac{mv}{\frac{h}{2\pi}}$$

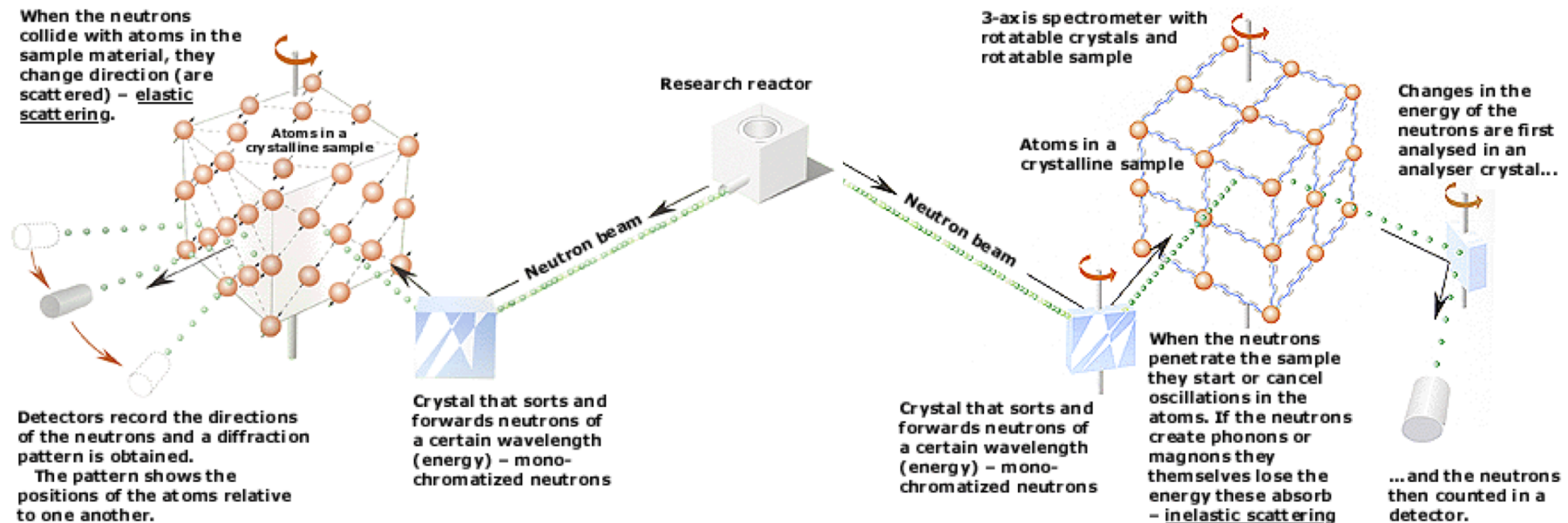
	Energy (meV)	Temperature (K)	Wavelength (Å)
cold	0.1 – 10	1 – 120	4 – 30
thermal	5 – 100	60 – 1000	1 – 4
hot	100 – 500	1000 – 6000	0.4 – 1

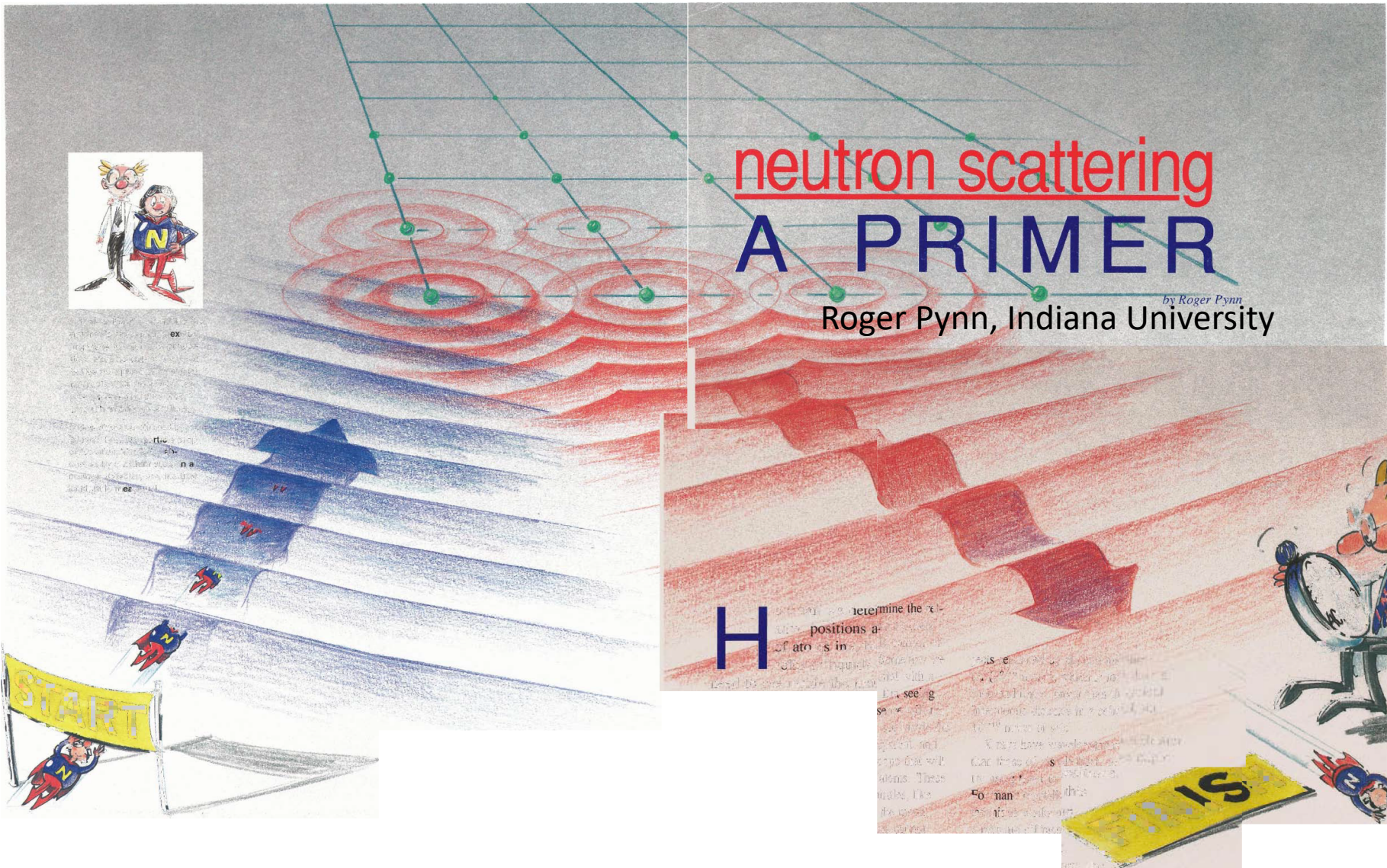
Nobel prize 1994 to Shull and Brockhouse

Neutrons see

Where atoms are

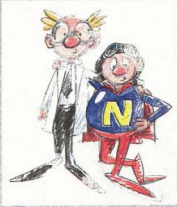
How atoms move





neutron scattering A PRIMER

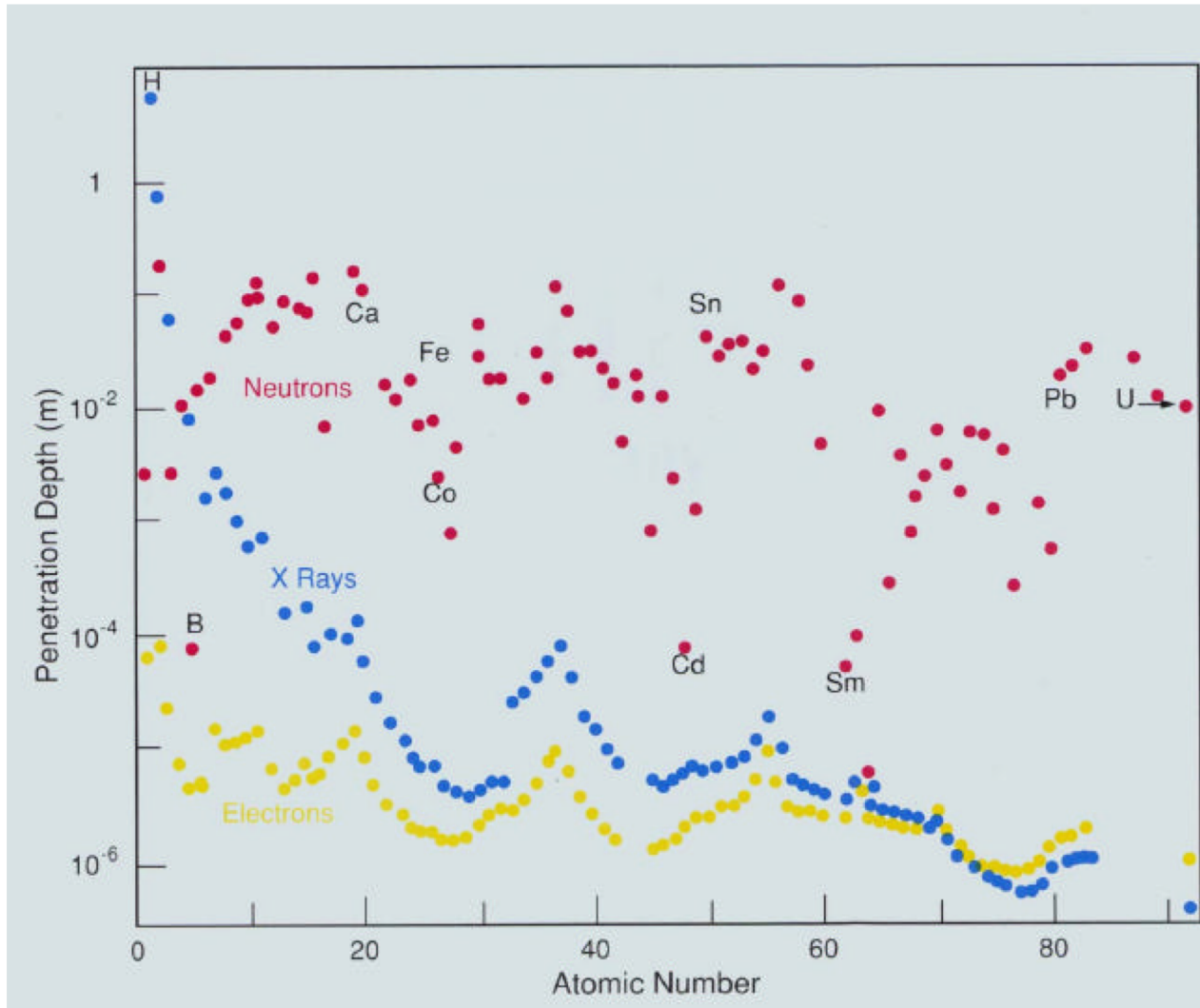
by Roger Pynn
Roger Pynn, Indiana University



Why we use neutrons

- Advantages:
 - **Wavelength** is in the order of **atomic distances** ($\approx 1\text{\AA} = 10^{-10}\text{ m}$)
 - **Energy** is in the order of **the kinetic energy of atoms** ($\approx \text{meV}$) and much smaller as the binding energy ($\approx \text{eV}$)
 - **Large penetration depth since uncharged particles**
 - Scattering is dependent on the **isotopic composition (Difference H,D)**
 - Neutrons have a magnetic Moment, they “**see**” B-fields
- Disadvantages:
 - Neutron sources have a very low Brilliance
 - Neutrons are difficult to detect, guide and to shield

Comparison of different probes

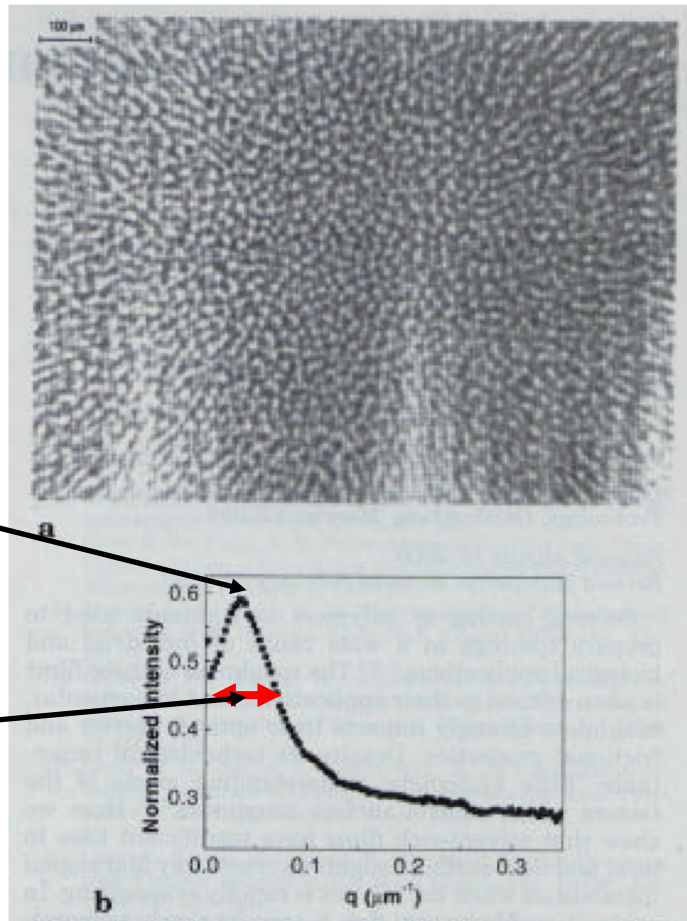


Neutrons:

- no systematic **A-dependence**
- Specific strongly absorbing isotopes: **B, Cd, Sm, Gd**
- Large difference for **H/D**

Scattering versus imaging measurements

- **Imaging techniques** are done in **real space**, like using a microscope
- Scattering techniques work on an **ensemble of objects in reciprocal space**
- Both methods are **complementary**

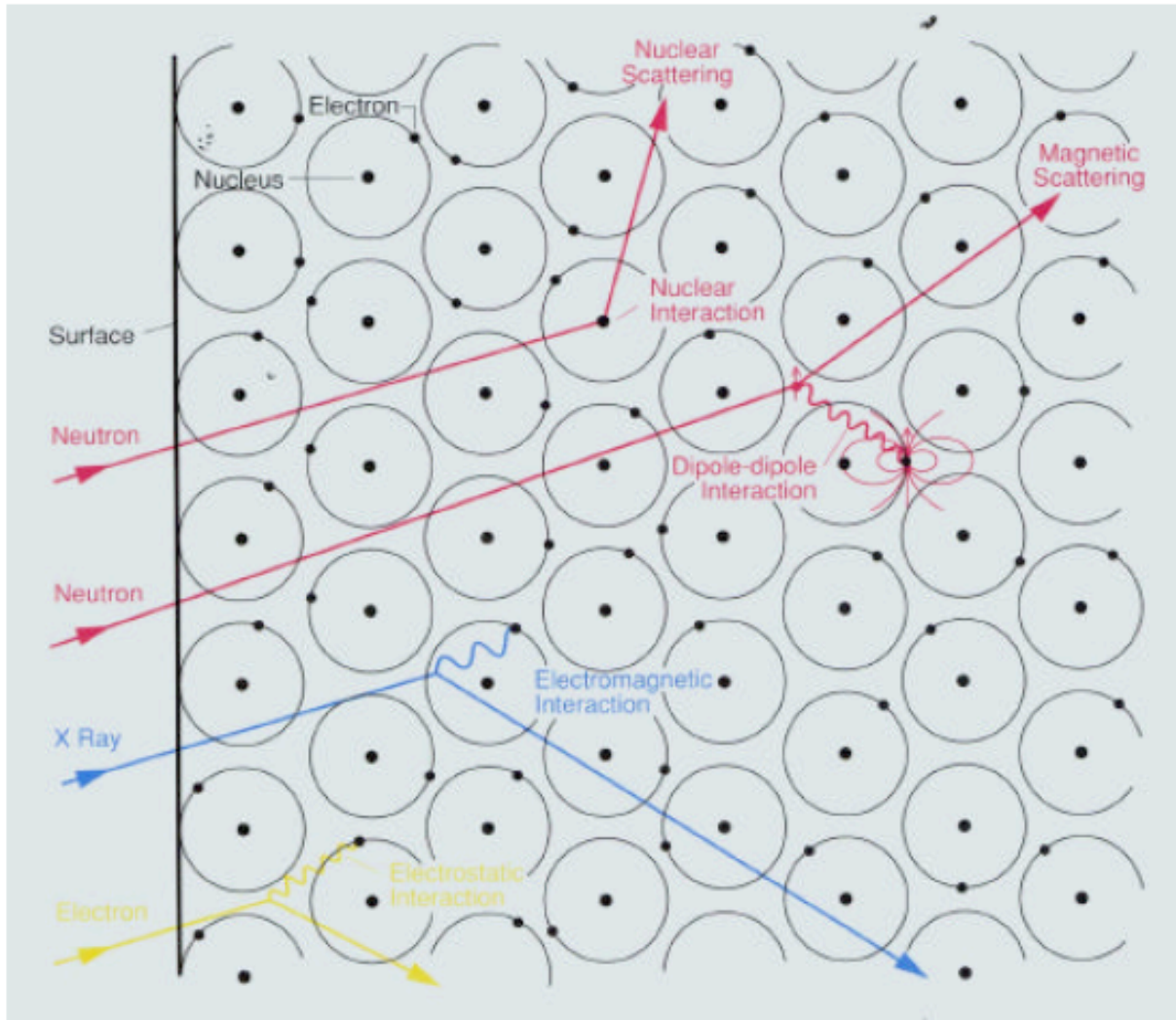


Foam in the sub micrometre range:
Picture from a microscope

Average size

Standard variation

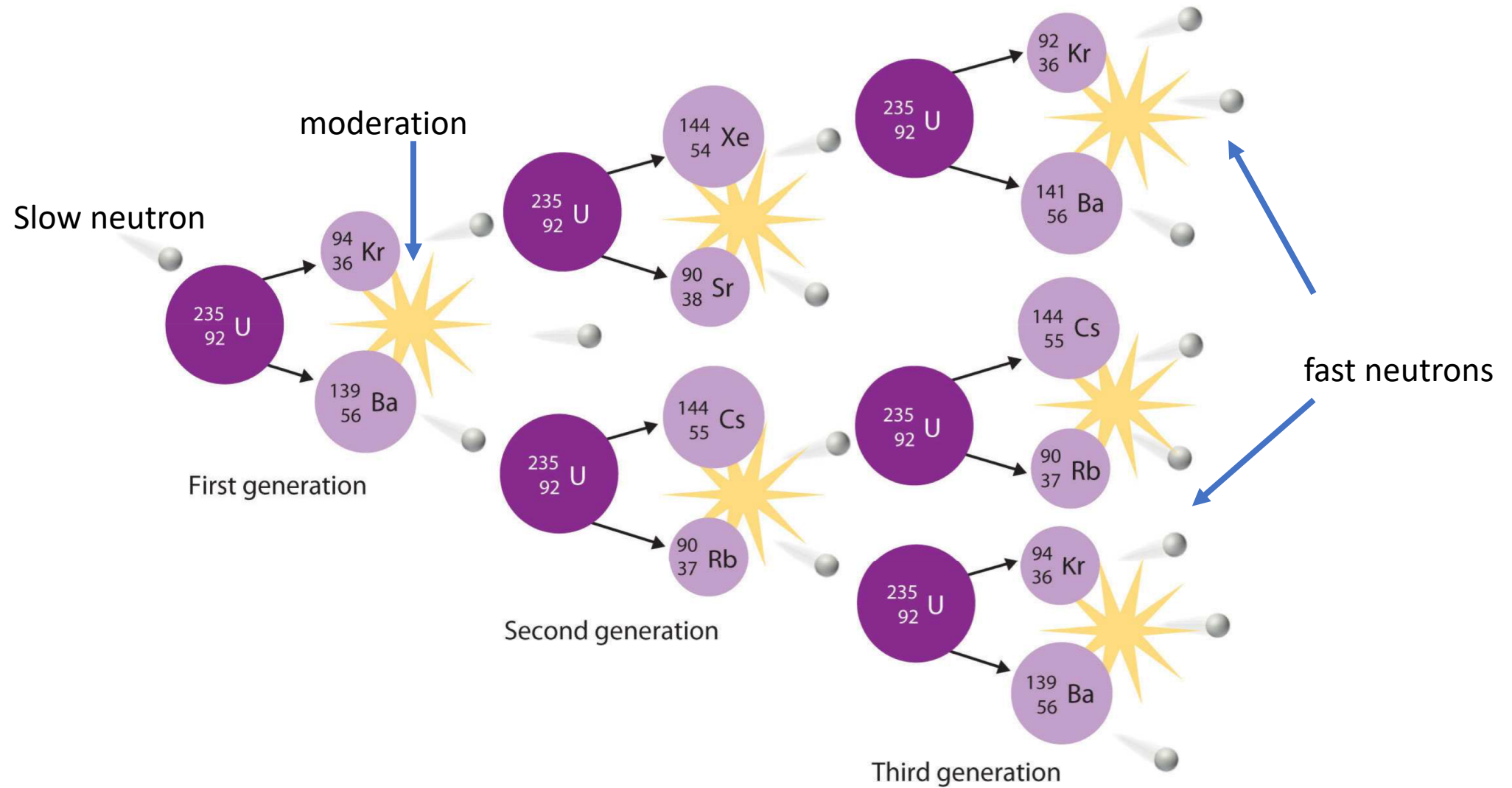
Interaction mechanism



Interaction of neutrons

- only with the **nucleus** (point interaction \sim fm)
- with **unpaired electrons** (**magnetic** Dipol-Dipol interaction)

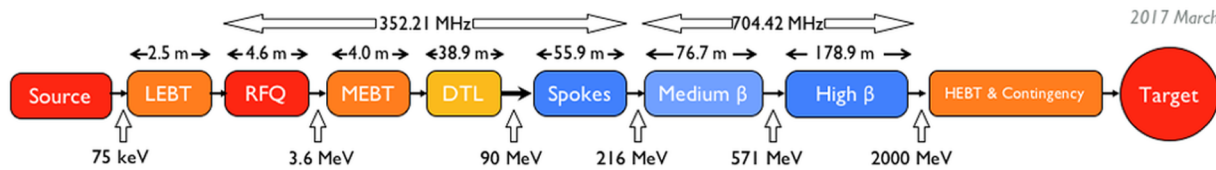
Fission: Chain reaction



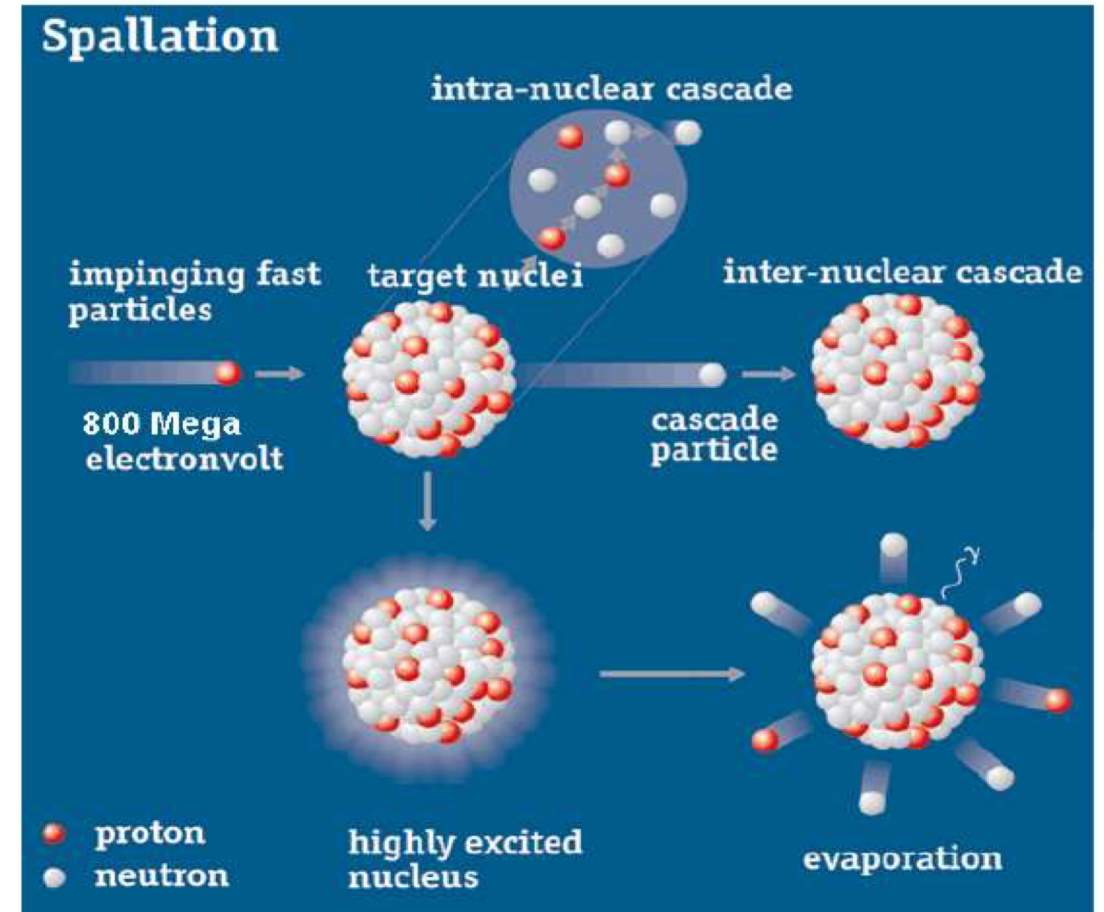
Spallation: Proton accelerator + heavy metal target

ESS: The world most powerful accelerator for the highest neutron flux

Accelerator for protons: 2 GeV and 5 mA



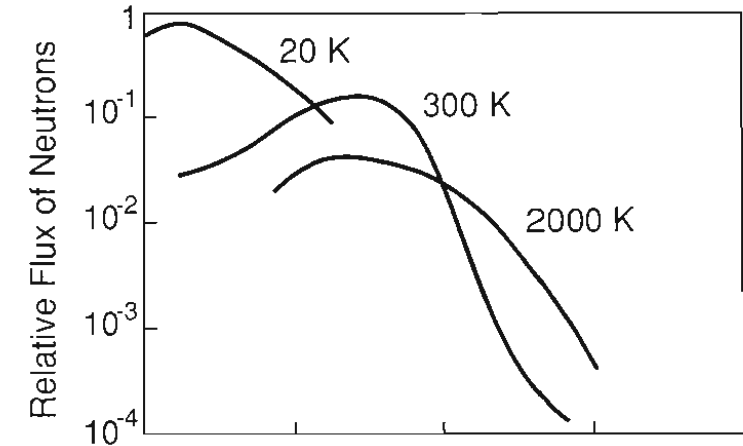
Tungsten target, He cooled



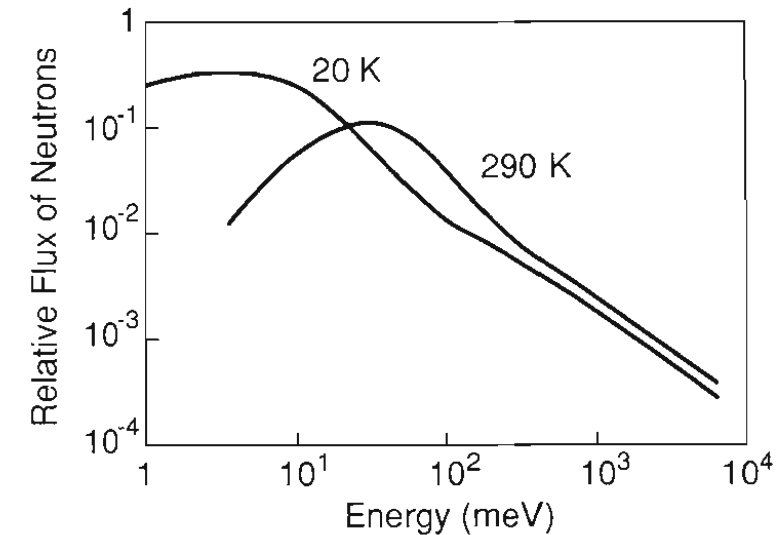
Fission versus Spallation

	Fission	Spallation
Energy per neutron	180 MeV	20 MeV
Neutron spectrum	Maxwellian	long tail of hot neutrons
Wavelength resolution	can be adopted to needs	constant
Time structure	continuous	pulsed
Stability	very stable	depended on accelerator
Problems	Nuclear reactor	much higher n energies
Building costs	About 0.5 Billion €	About 2 Billion €
Running costs	Current for pumps	Current for accelerator
Further improvements	Saturation reached	Higher accelerator energy

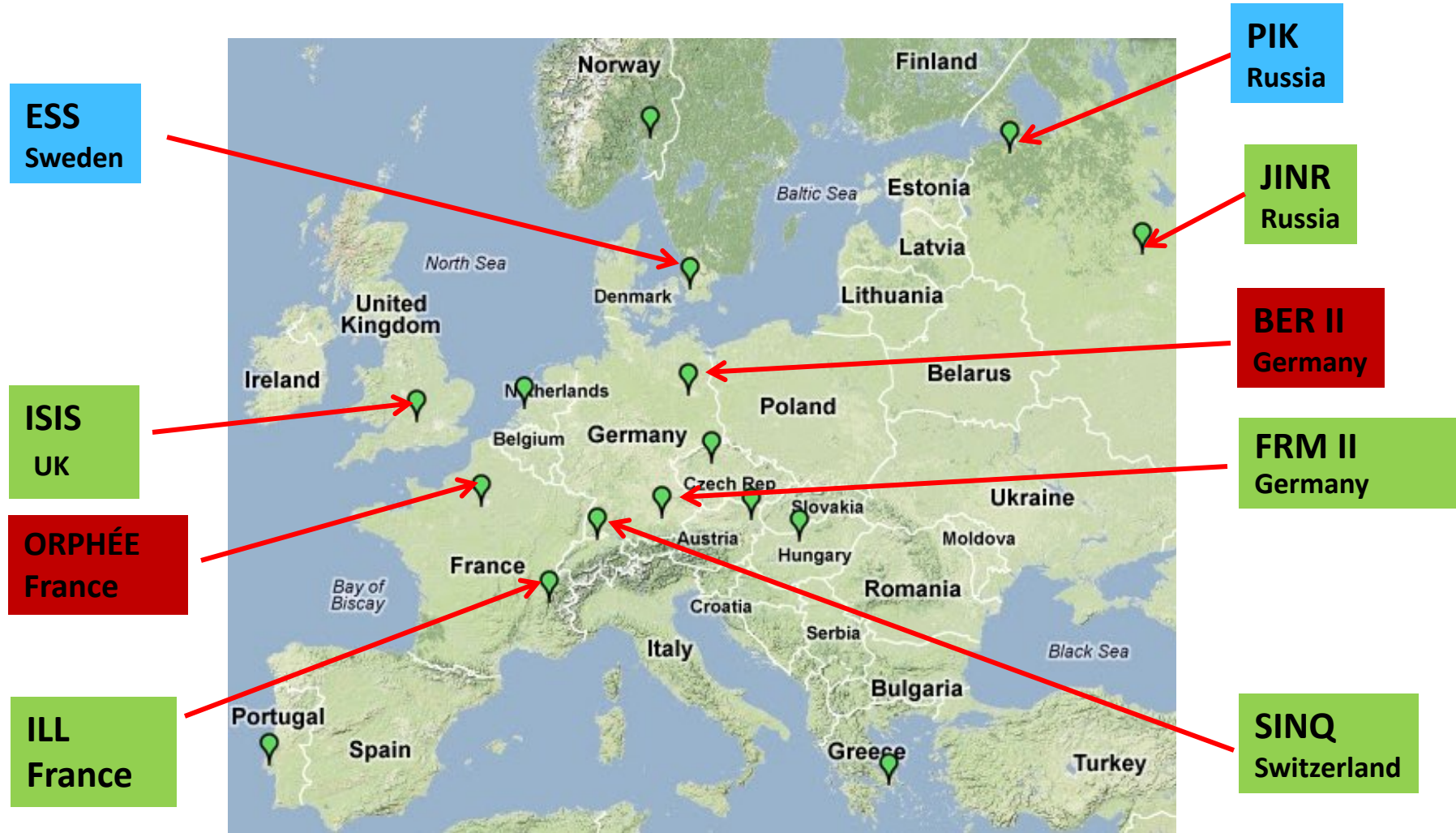
Reactor spectrum



Spallation spectrum



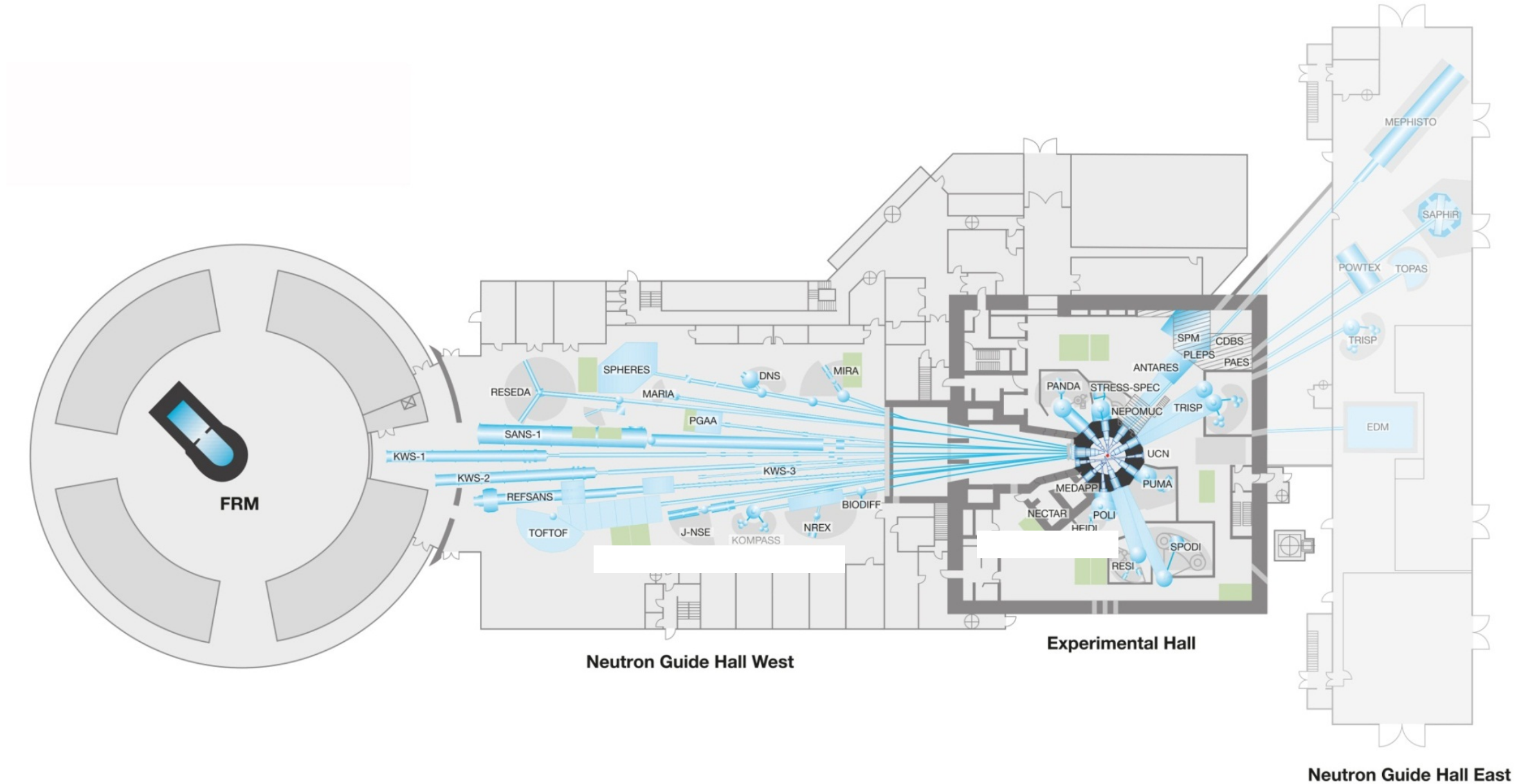
European Landscape of Neutron User Facilities



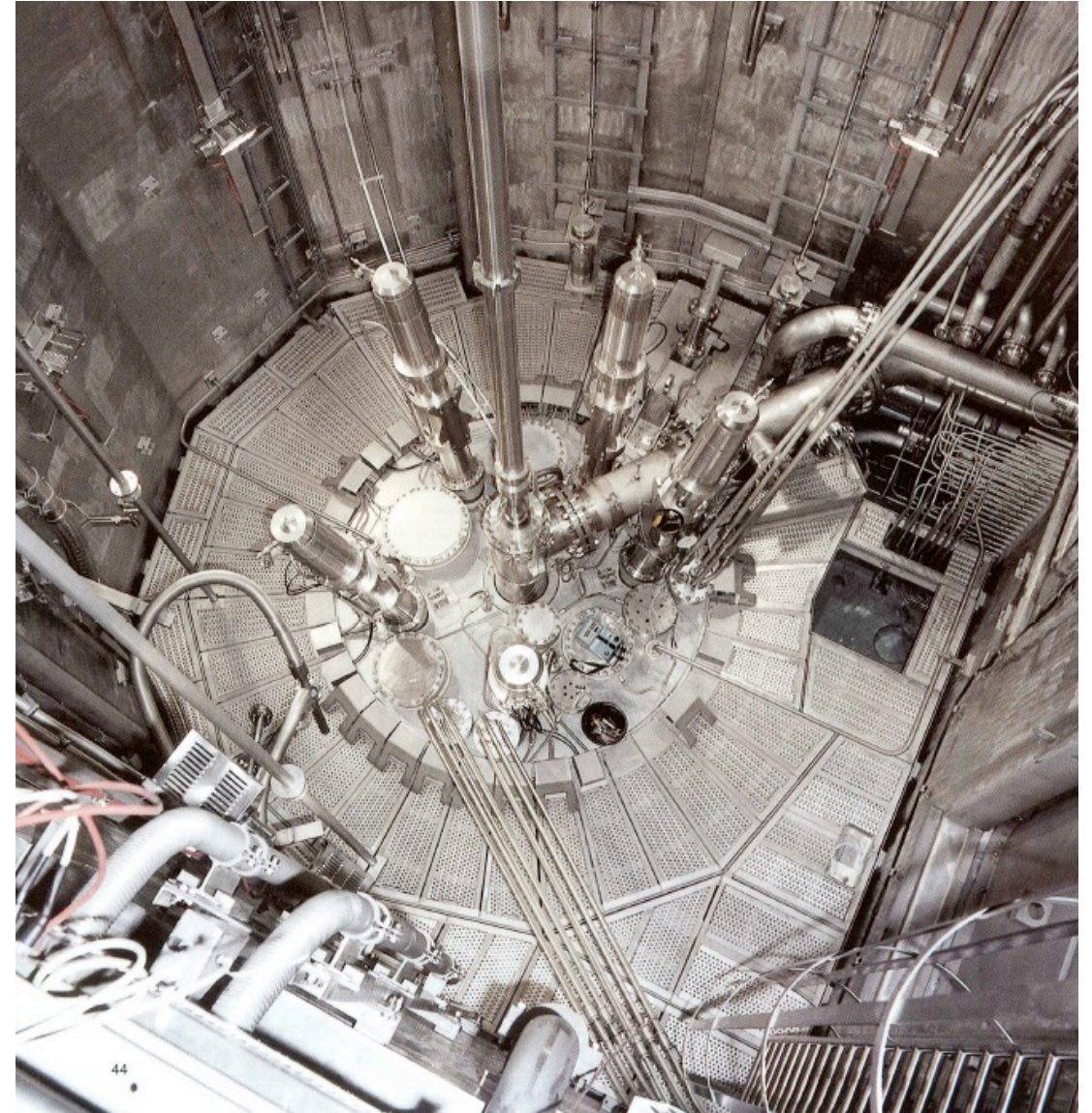
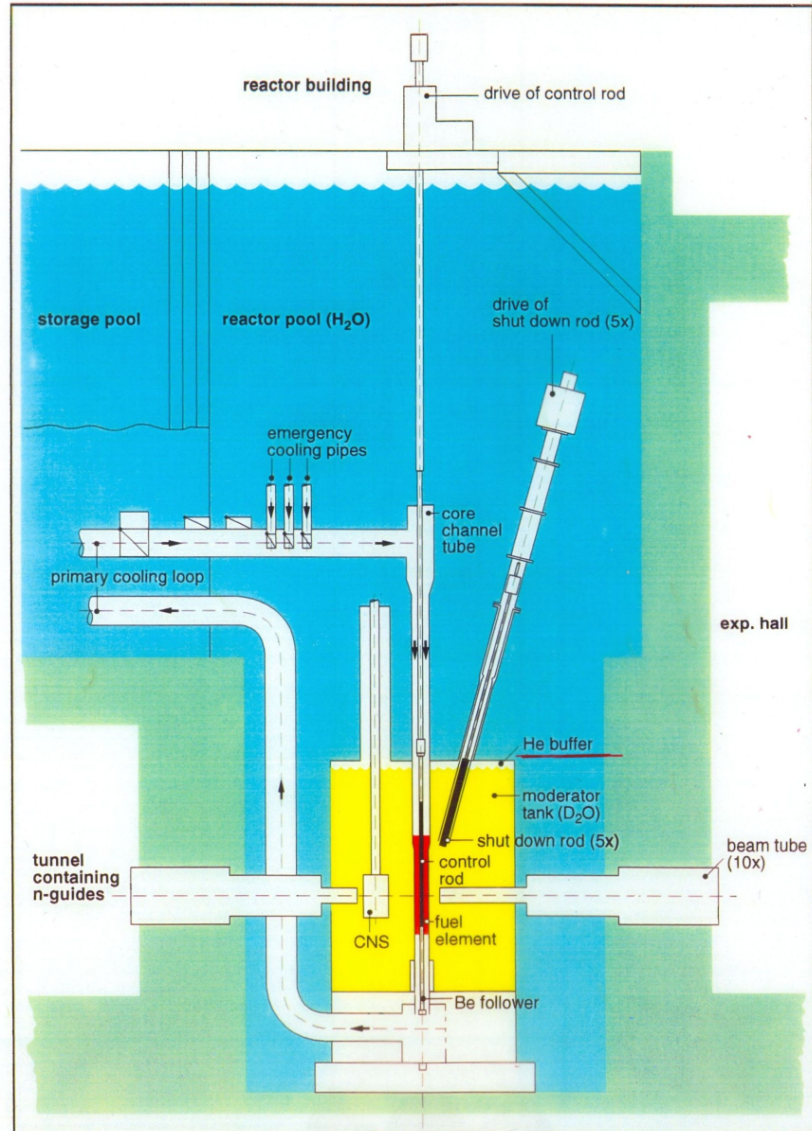
International Neutron Sources

	ILL Grenoble (F)	IBR Dubna (Russia)	ISIS Chilton (GB)	FRM II	ESS Lund (S)	SNS Oak Ridge (USA)
Φ [$cm^{-2}s^{-1}$]	10^{15}	2×10^{16}	4.5×10^{15}	7×10^{14}	1.5×10^{17}	8×10^{16}
$\bar{\Phi}$ [$cm^{-2}s^{-1}$]	10^{15}	2×10^{13}	7×10^{12}	7×10^{14}	0.6×10^{15}	6×10^{13}
Pulse repetition rate [Hz]	-	5	50	-	14	60
Pulse duration [μs]	-	250	30	-	2860	20
P [MW]	57	2	0.2	20	5	2

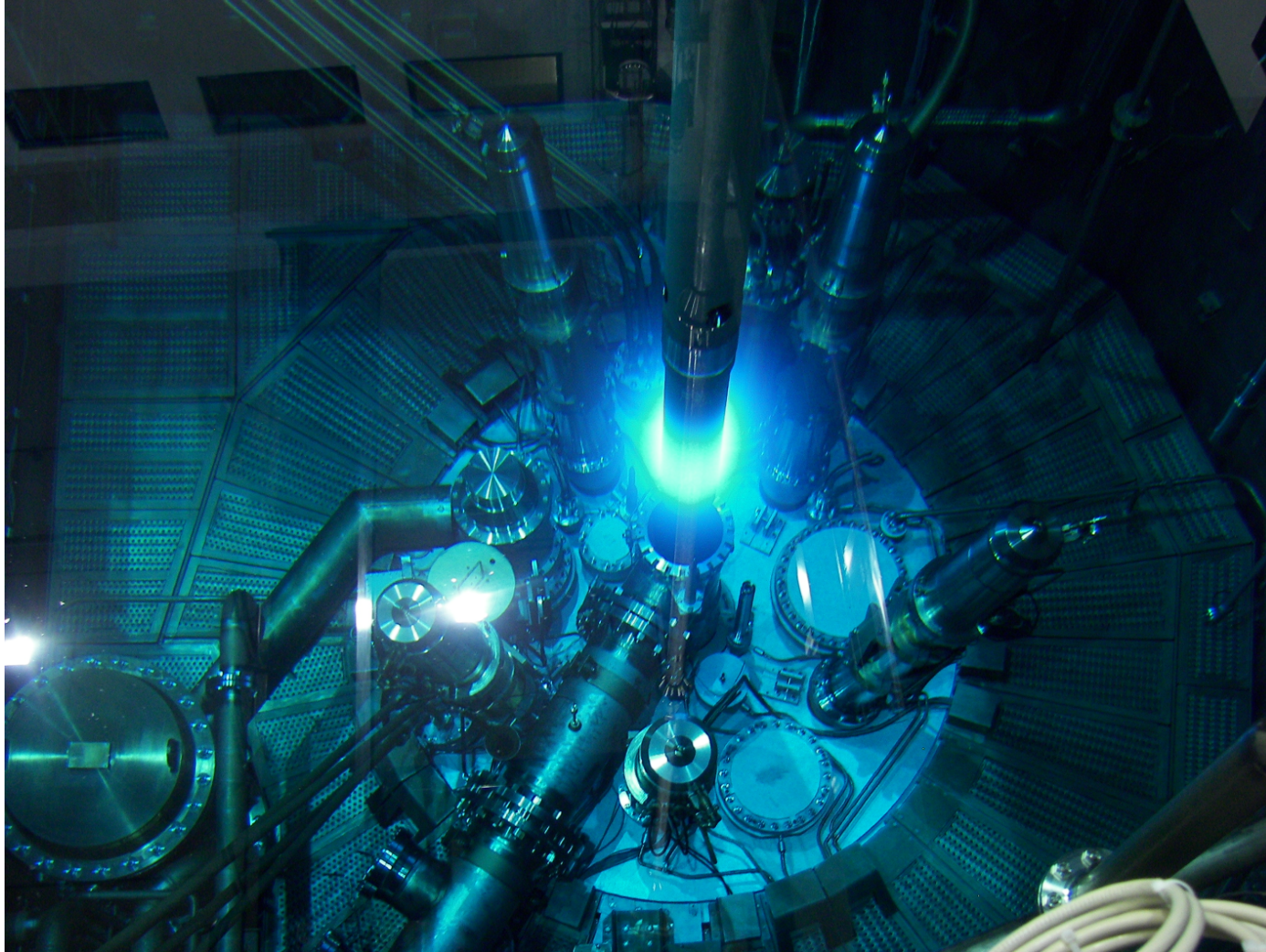
The Neutron Source FRM II



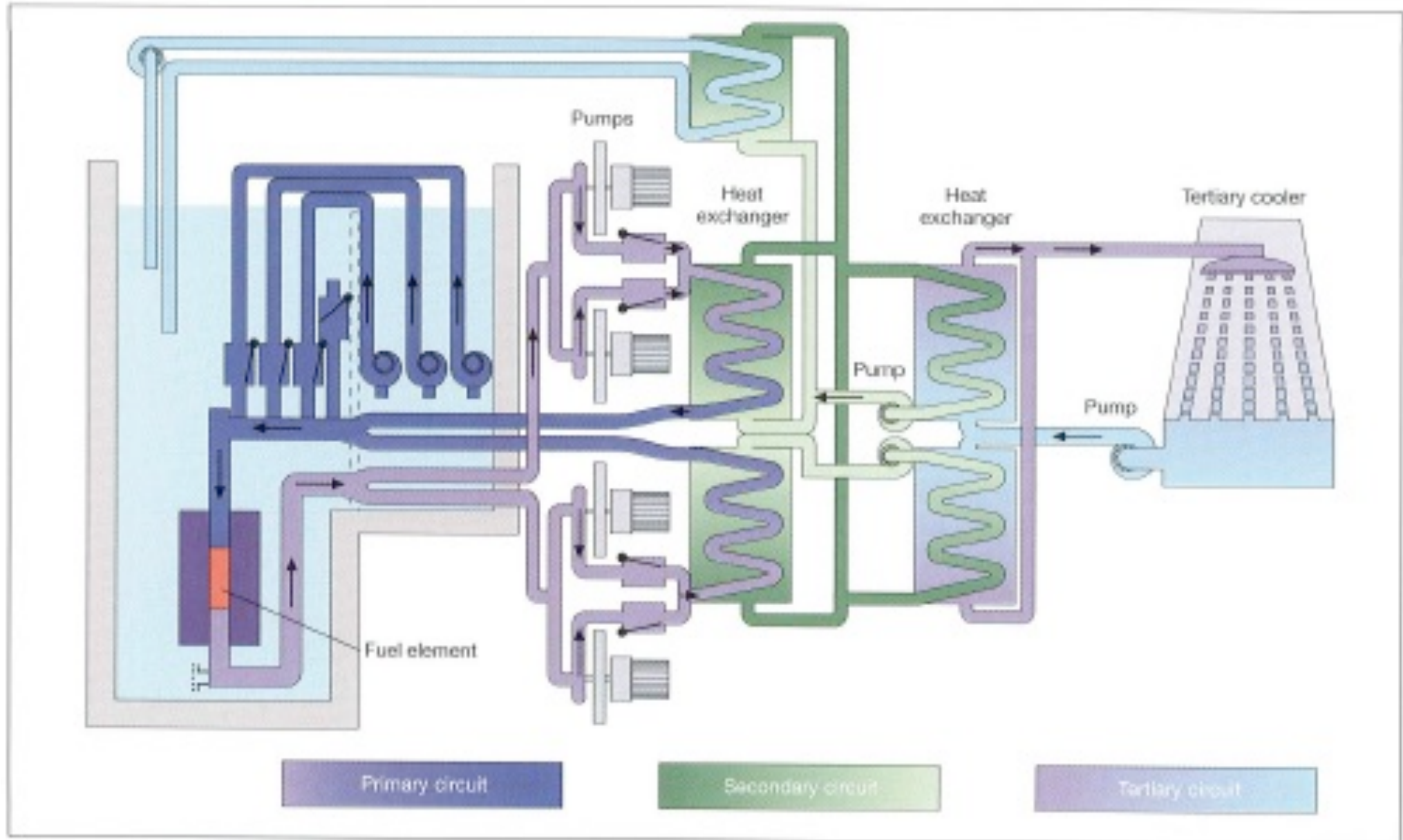
FRM II: A swimming pool reactor



FRM II: Discharge of a spent fuel element

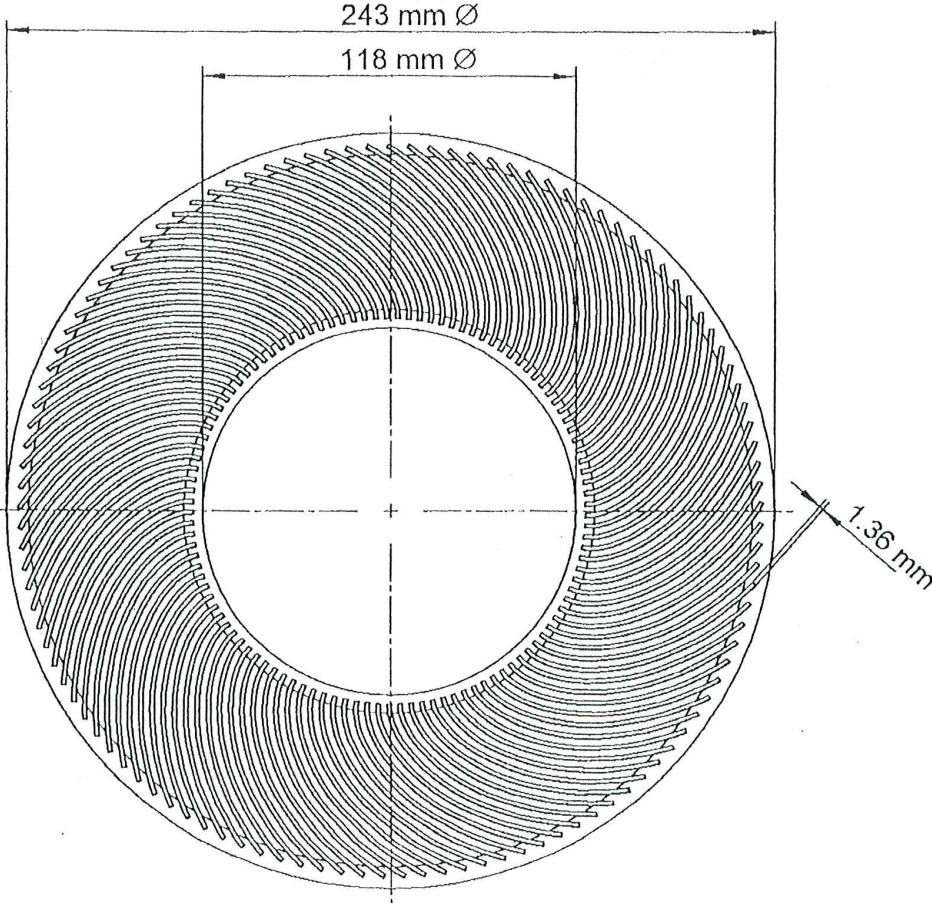


FRM II: Cooling system

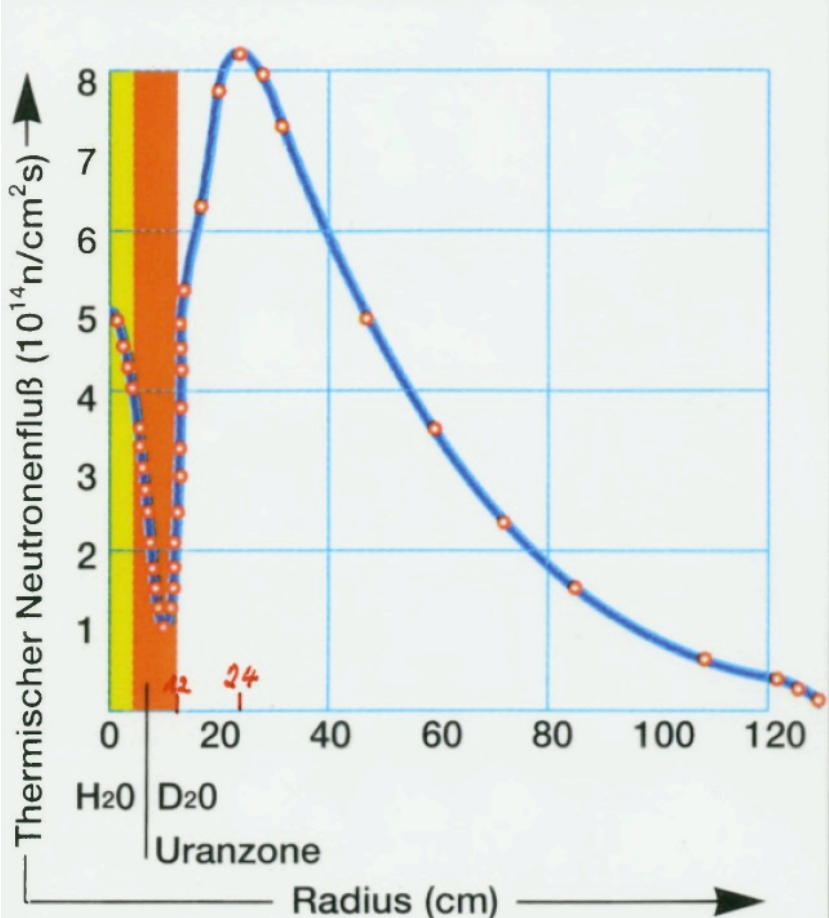


FRM II: The Fuel Element

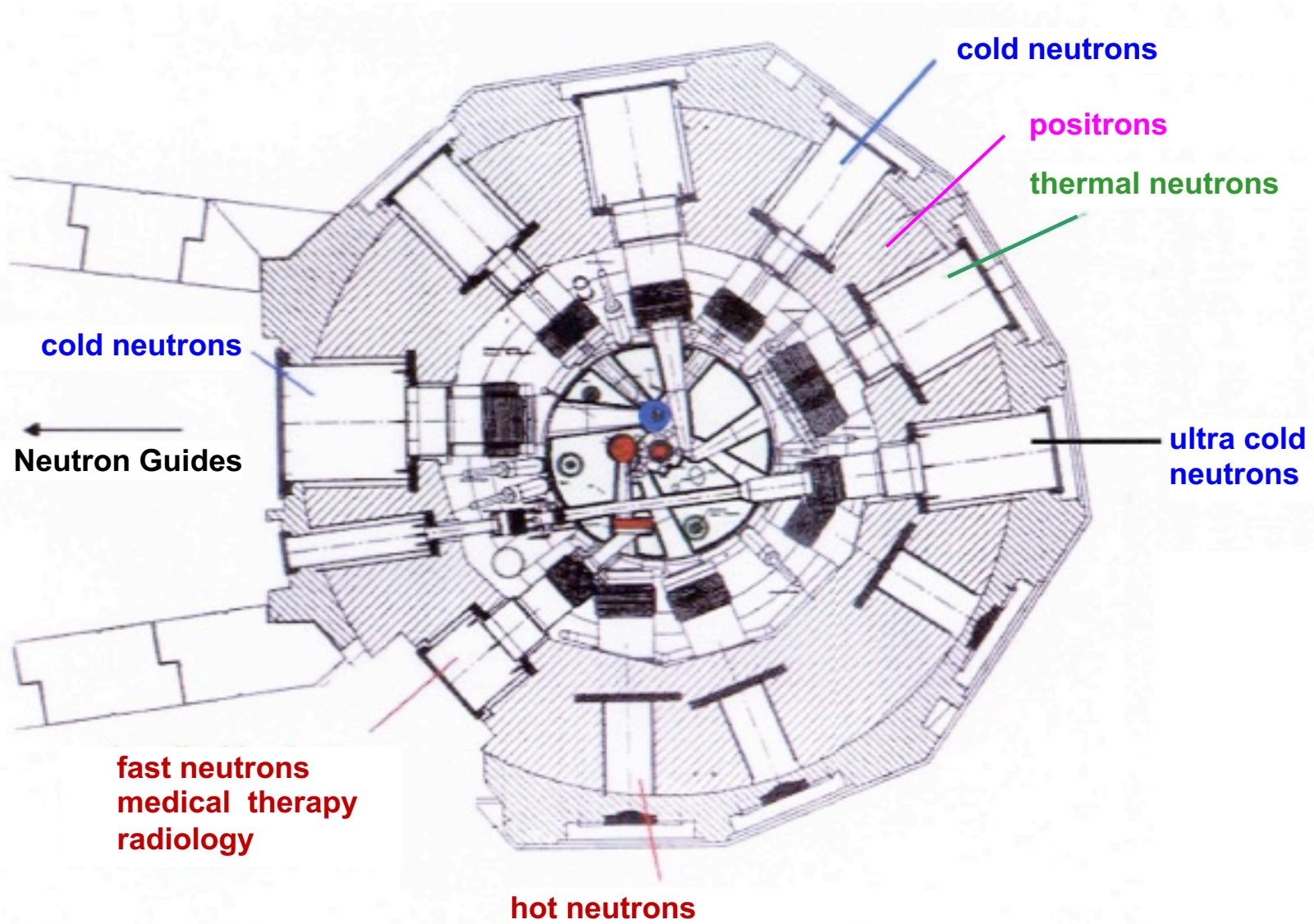
Cross section: 113 curved fuel plates



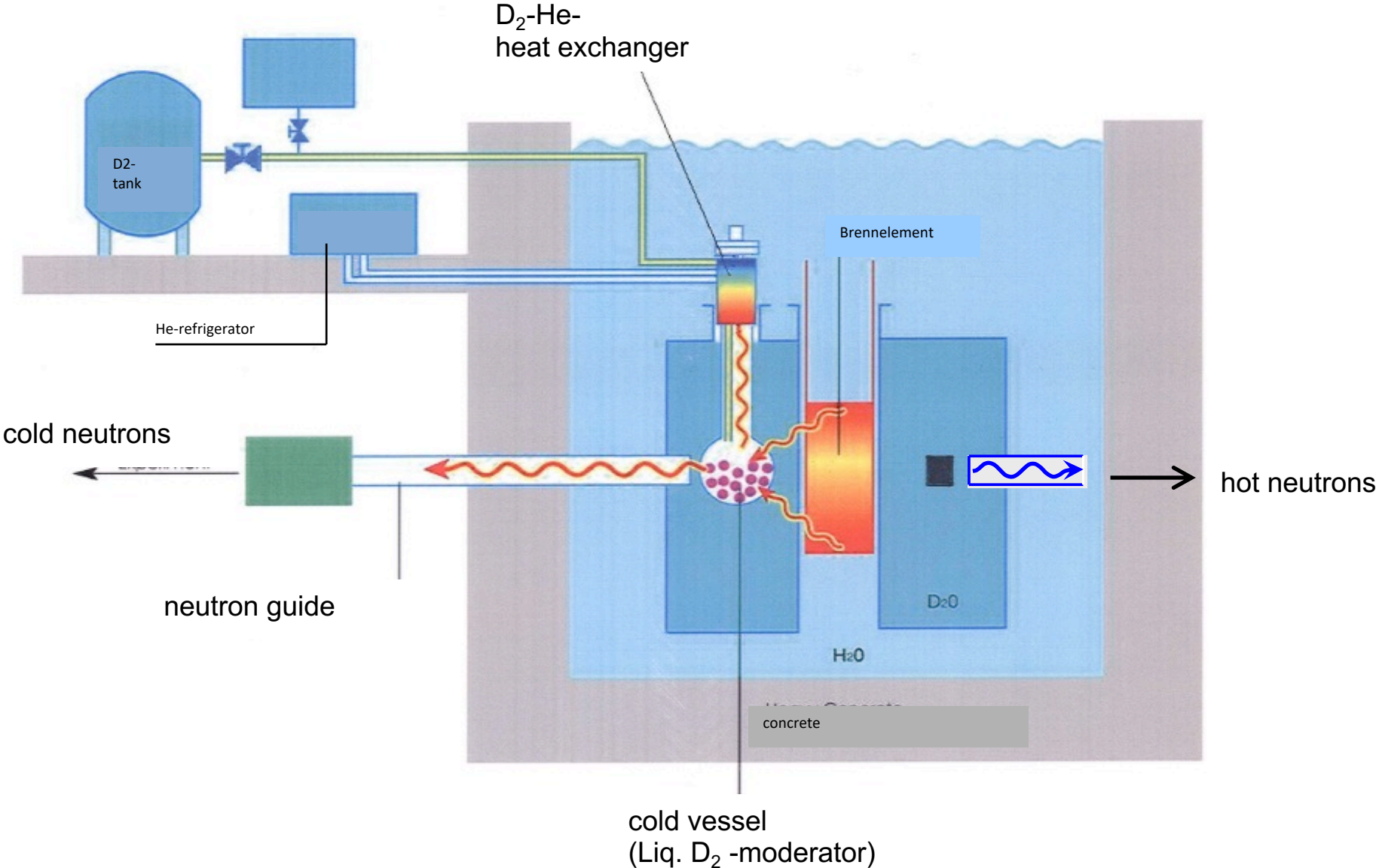
Flux maximum 12 cm above fuel element



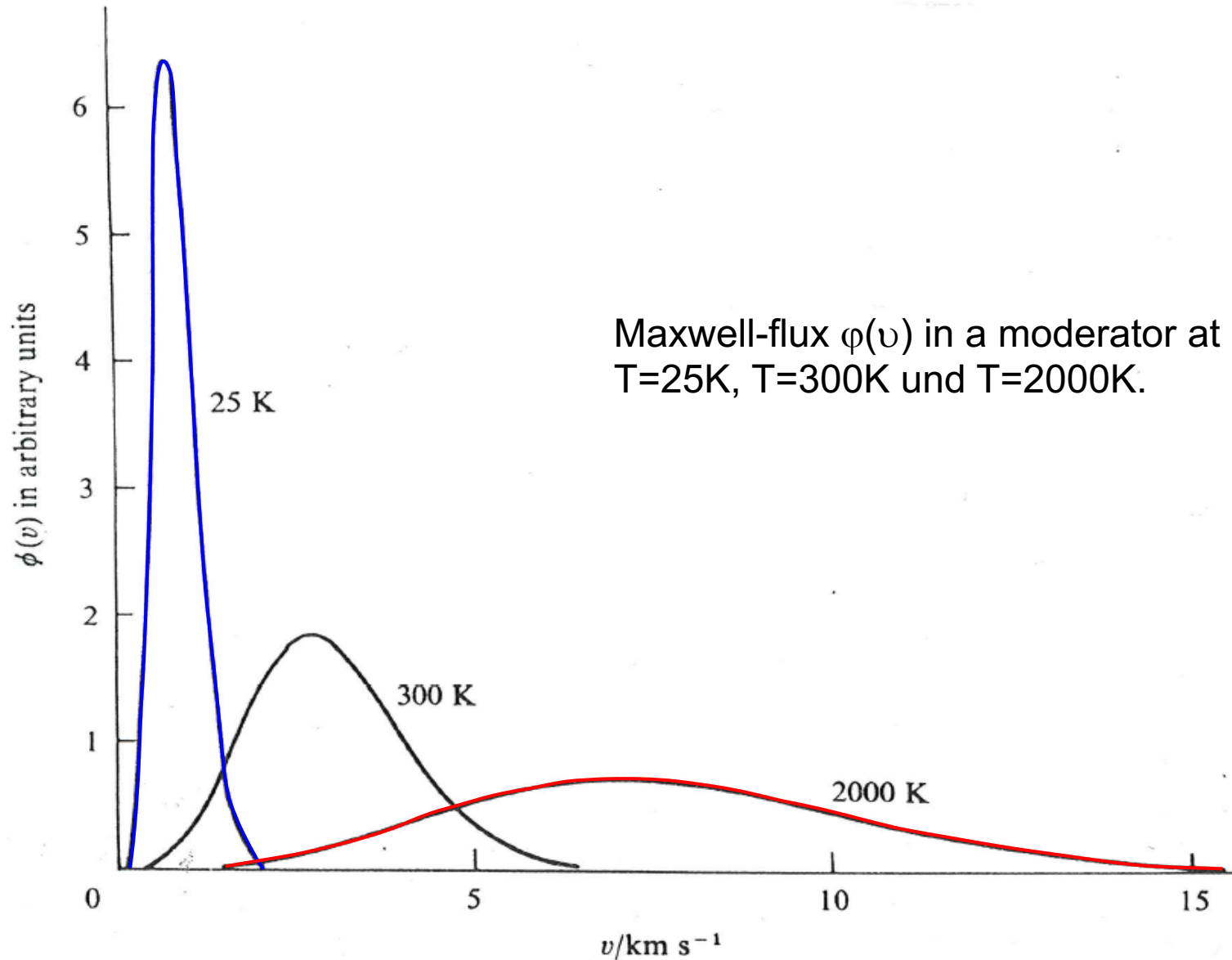
FRM II: The reactor vessel



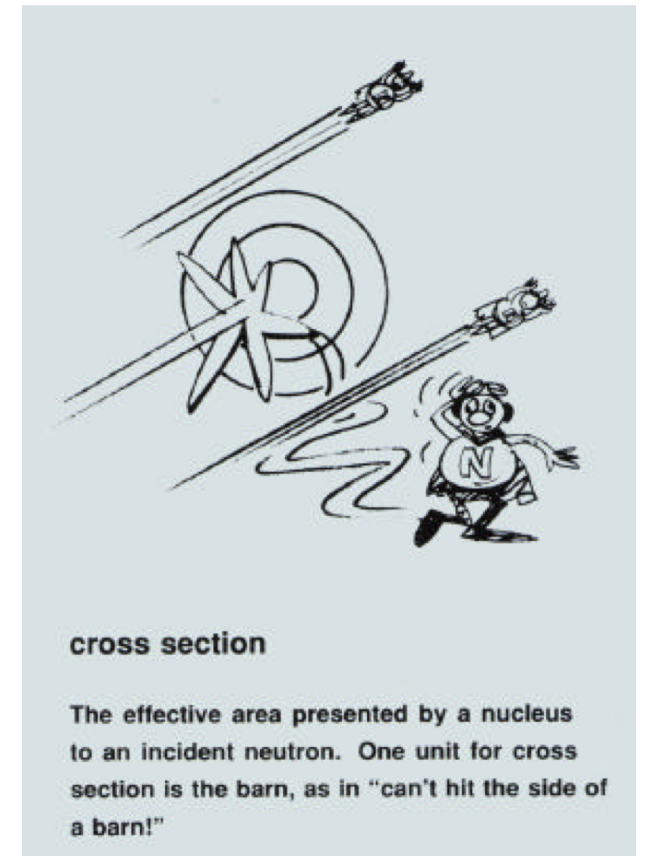
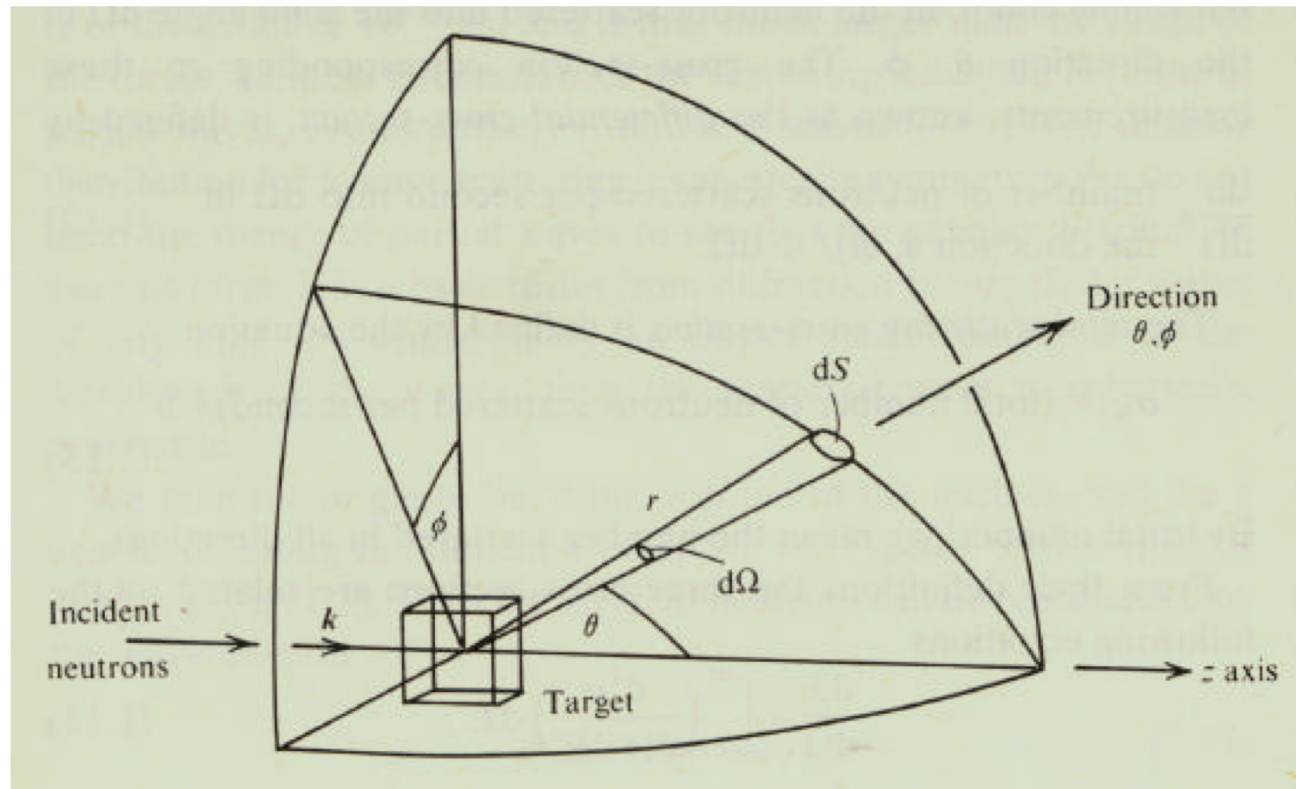
FRM II: Cold Source, Hot Source



FRM II: Spectrum of the cold source and hot source



Scattering theory: Cross section

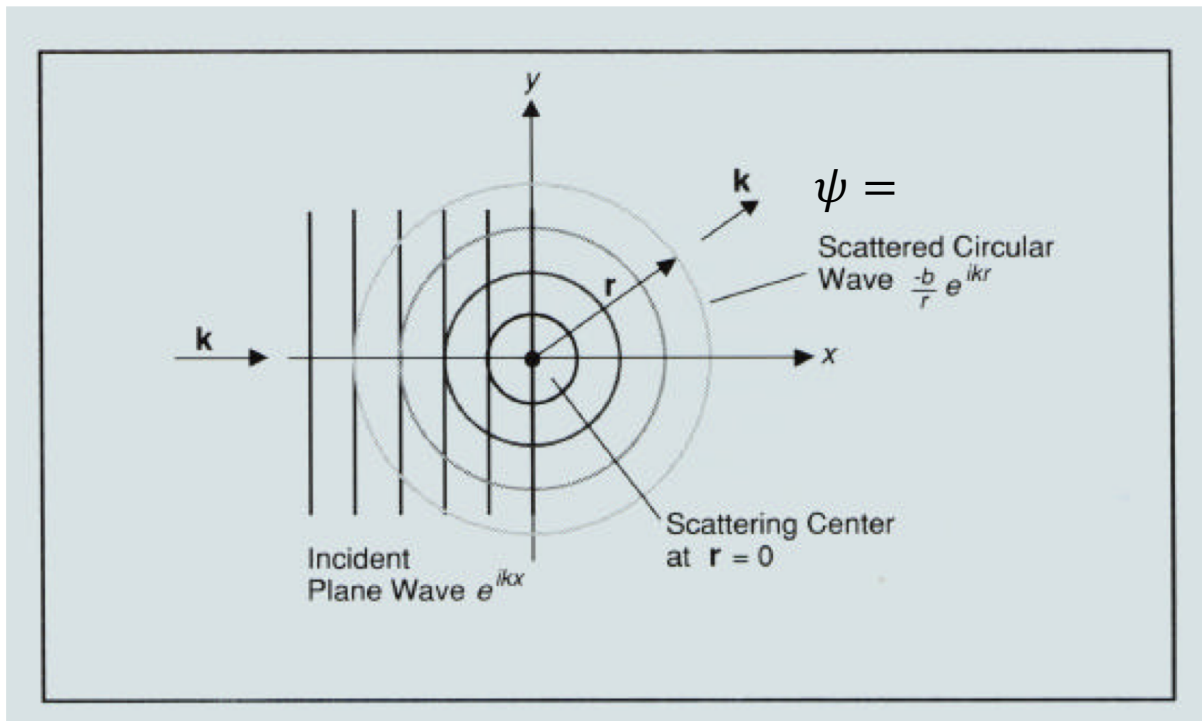


- Flux Φ = number of incident $n/(s \text{ cm}^2)$
- Cross section σ = number of scattered $n/s / \Phi$
- $d\sigma/d\Omega$ = number of scattered $n/s / \Phi d\Omega$

$$\text{Attenuation} = e^{-N\sigma d}$$

σ in barn: 1 barn = 10^{-24} cm^2
 N = Atoms/unit cell
 d = thickness

Scattering on a single nucleus



- Strong interaction short range ($\sim 1\text{fm}$) \ll neutron wavelength
 \Rightarrow scattering is “point like”
- no absorption
- elastic (no energy transfer, no time dependency)
- b is the scattering length in cm, typical 10^{-12} cm (can not be calculated, needs to be measured !)

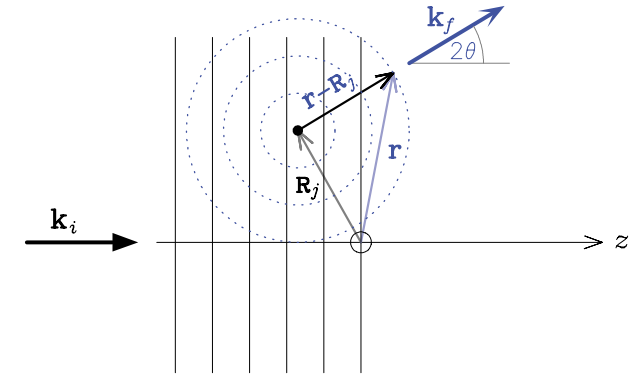
• Differential cross section:
$$\frac{d\sigma}{d\Omega} = \frac{\text{number of scatter } n/s}{d\Omega \Phi} = \frac{|\psi|^2}{d\Omega \Phi} = b^2$$

• Total cross section:
$$\sigma = 4\pi b^2$$

Coherent and incoherent scattering

- Superposition of all neutron waves: sum over all atoms N:

$$\psi_{scatter} = \sum_{i,j=0\dots N} e^{-i\vec{k}_0 \cdot \vec{R}_i} \left[\frac{-b_j}{|\vec{r} - \vec{R}_j|} e^{-i\vec{k}_f \cdot (\vec{r} - \vec{R}_j)} \right]$$



- For neutrons the the scattering length depends on the isotope and the nuclear spin: $b_i = \langle b \rangle + \delta b_i$
- This gives in the end

$$\frac{d\sigma}{d\Omega} = \langle b \rangle^2 \sum_{i,j} e^{-i\vec{Q} \cdot (\vec{R}_i - \vec{R}_j)} + (\langle b^2 \rangle - \langle b \rangle^2) N$$

coherent scattering
(**Q dependent**)

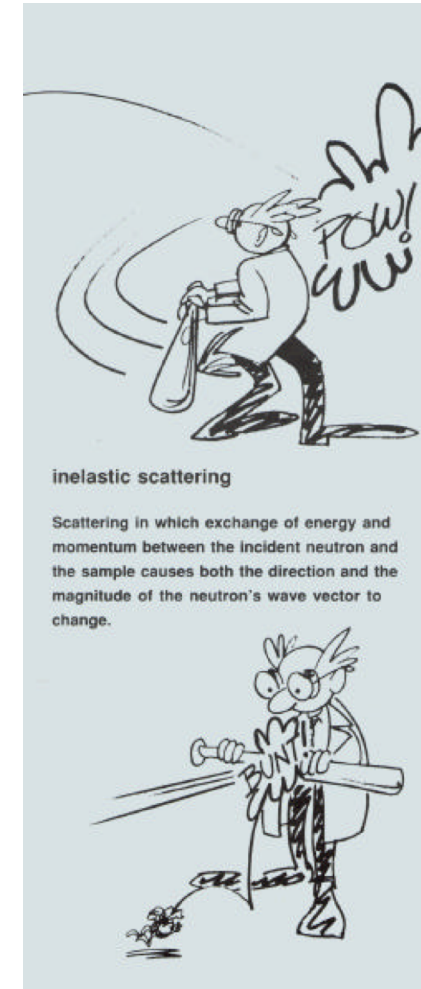
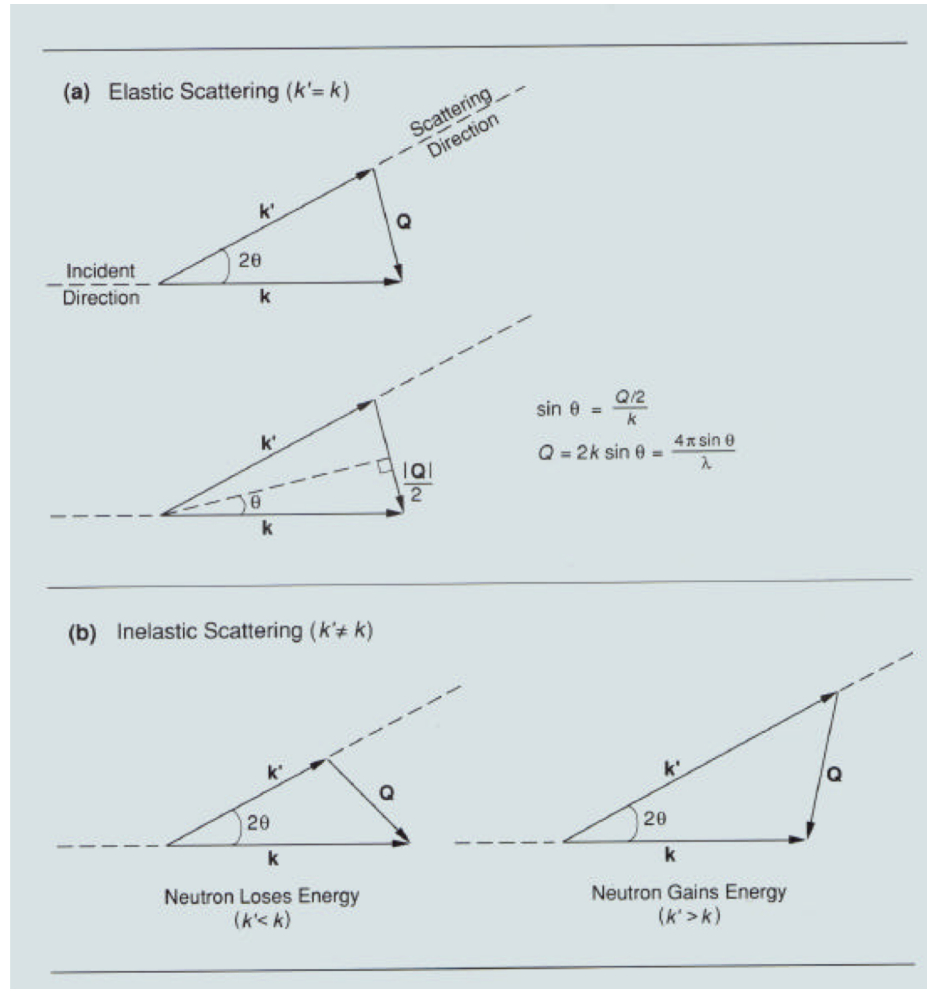
incoherent scattering
(**in all directions**)

Selected values for σ_{coh} and σ_{inc}

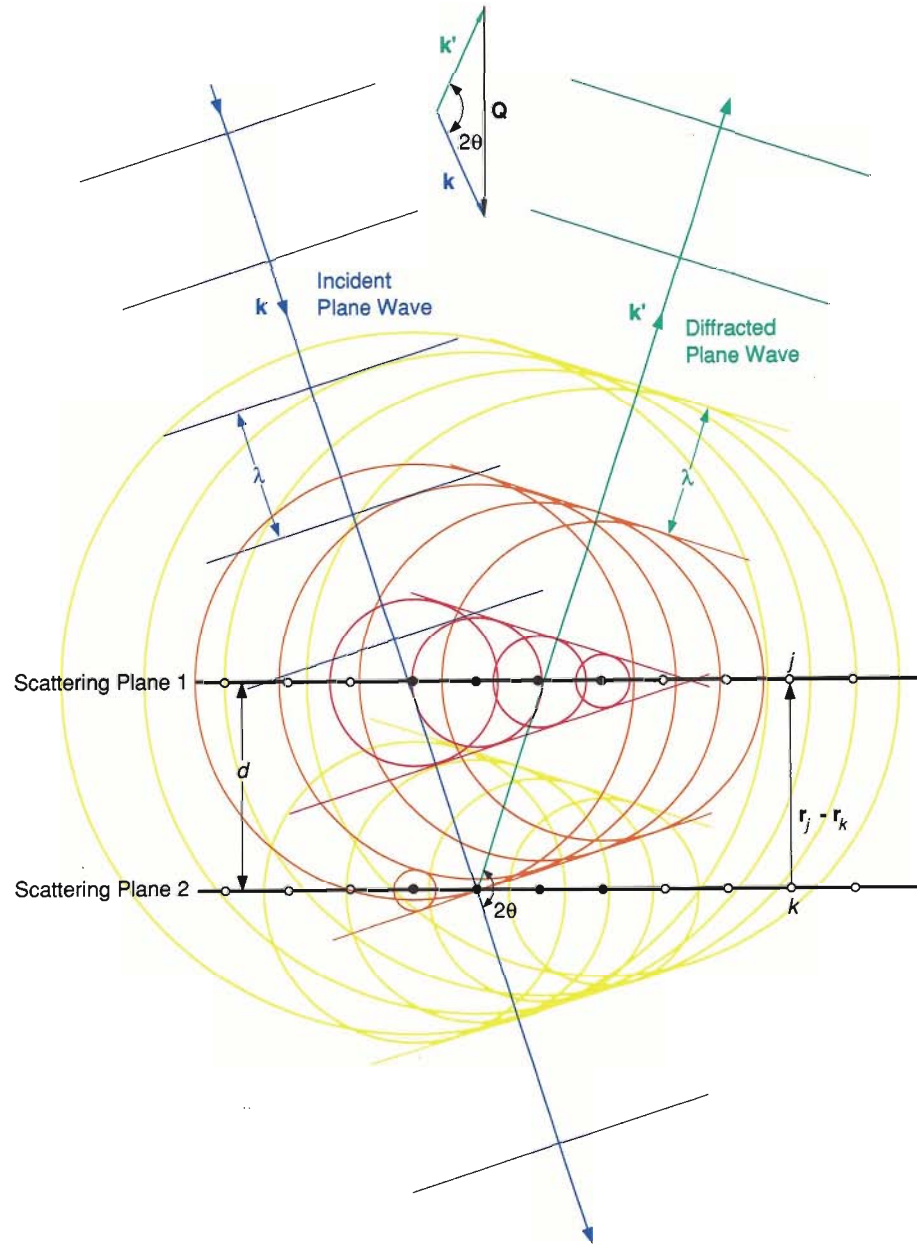
Nuclide	σ_{coh}	σ_{inc}	Nuclide	σ_{coh}	σ_{inc}
^1H	1.8	80.2	V	0.02	5.0
^2H	5.6	2.0	Fe	11.5	0.4
C	5.6	0.0	Co	1.0	5.2
O	4.2	0.0	Cu	7.5	0.5
Al	1.5	0.0	^{36}Ar	24.9	0.0

- Large difference for H/D which is used for contrast variation
- Al is used for sample environment and beam windows
- V is used as a standard scatter for inelastic scattering

Elastic versus inelastic scattering



Coherent elastic scattering at crystals



Condition for constructive interference:

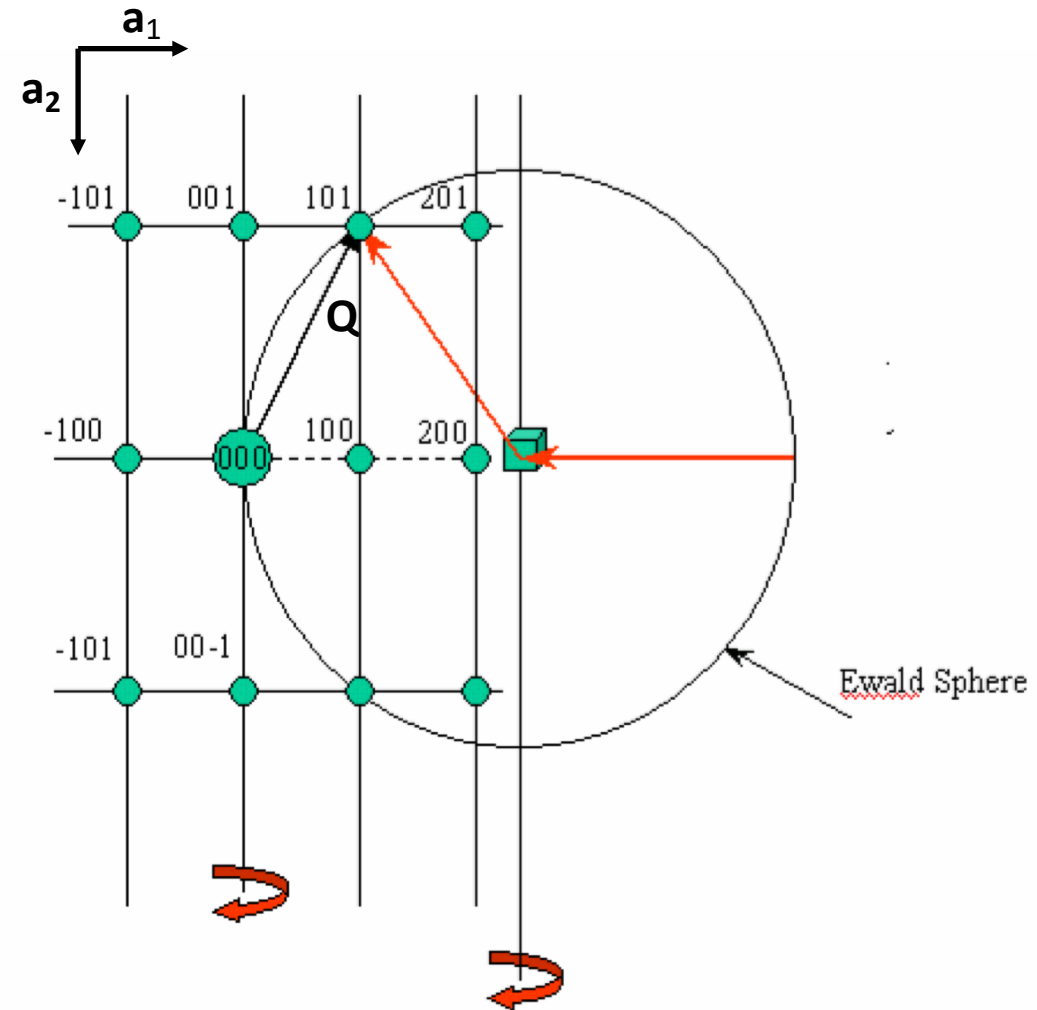
- \vec{Q} must be perpendicular to the diffracted wave front
- $\vec{Q} \cdot (\vec{r}_j - \vec{r}_k) = Qd = 2\pi n$, where d is the lattice spacing, and n is an integer number
- $Q = \frac{4\pi}{\lambda} \sin \theta$ is the condition for elastic scattering

\Rightarrow

$$n\lambda = 2d \sin \theta \quad \text{Bragg's law}$$

Key Points about Diffraction

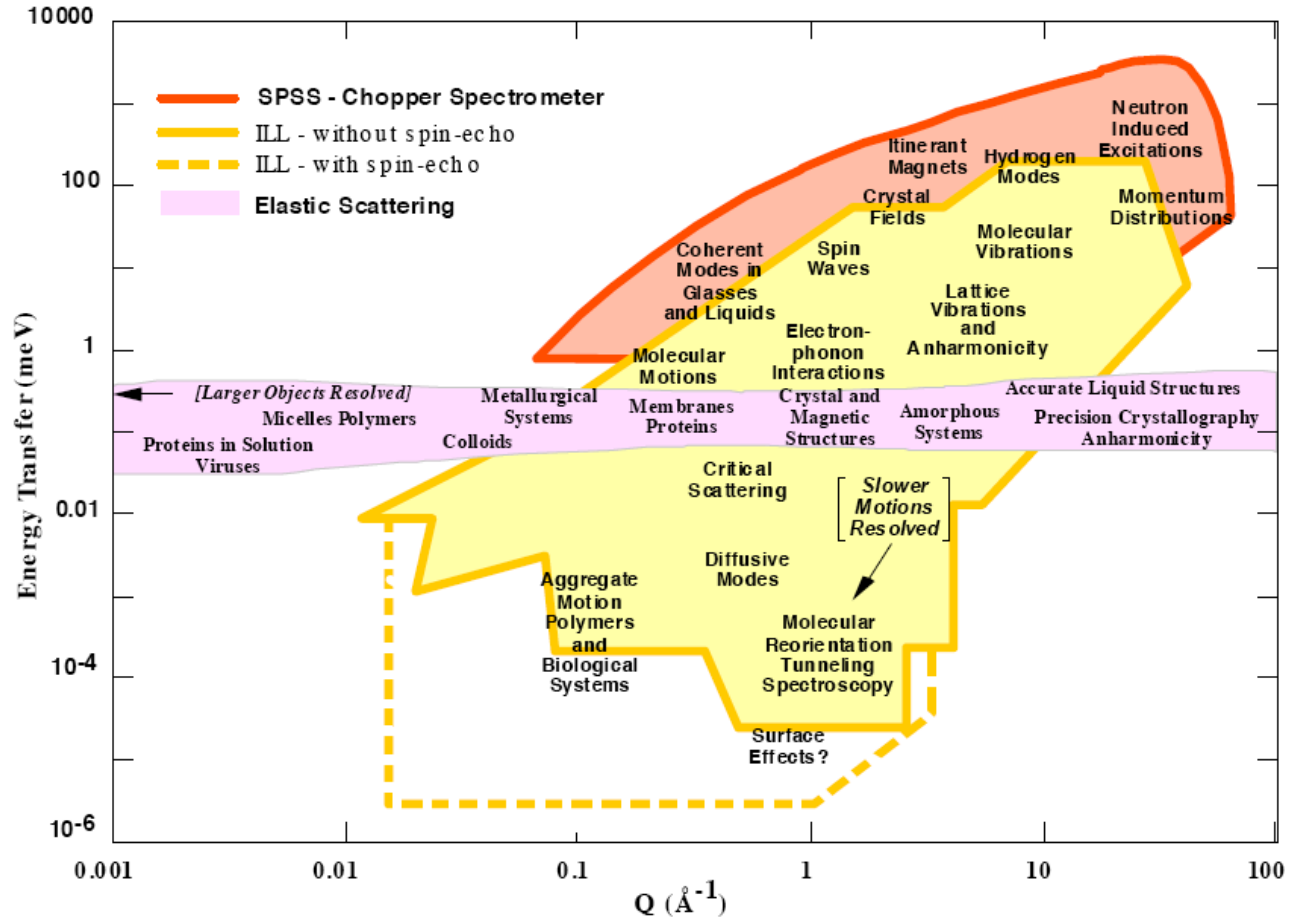
- A **monochromatic (single λ)** neutron beam is diffracted by a single crystal only if specific geometrical conditions are fulfilled
- These conditions can be expressed in several ways:
 - **Laue's conditions:** $\mathbf{Q} \cdot \mathbf{a}_1 = h$; $\mathbf{Q} \cdot \mathbf{a}_2 = k$; $\mathbf{Q} \cdot \mathbf{a}_3 = l$
h, k, and l as integers; \mathbf{a}_i the translations of the unit cell
 - **Bragg's Law:** $2d_{hkl}\sin\theta = \lambda$
 - Ewald's construction
- Diffraction tells us about:
 - The **dimensions** of the unit cell
 - The **symmetry** of the crystal
 - The **positions** of atoms within the unit cell
 - The extent of thermal vibrations of atoms



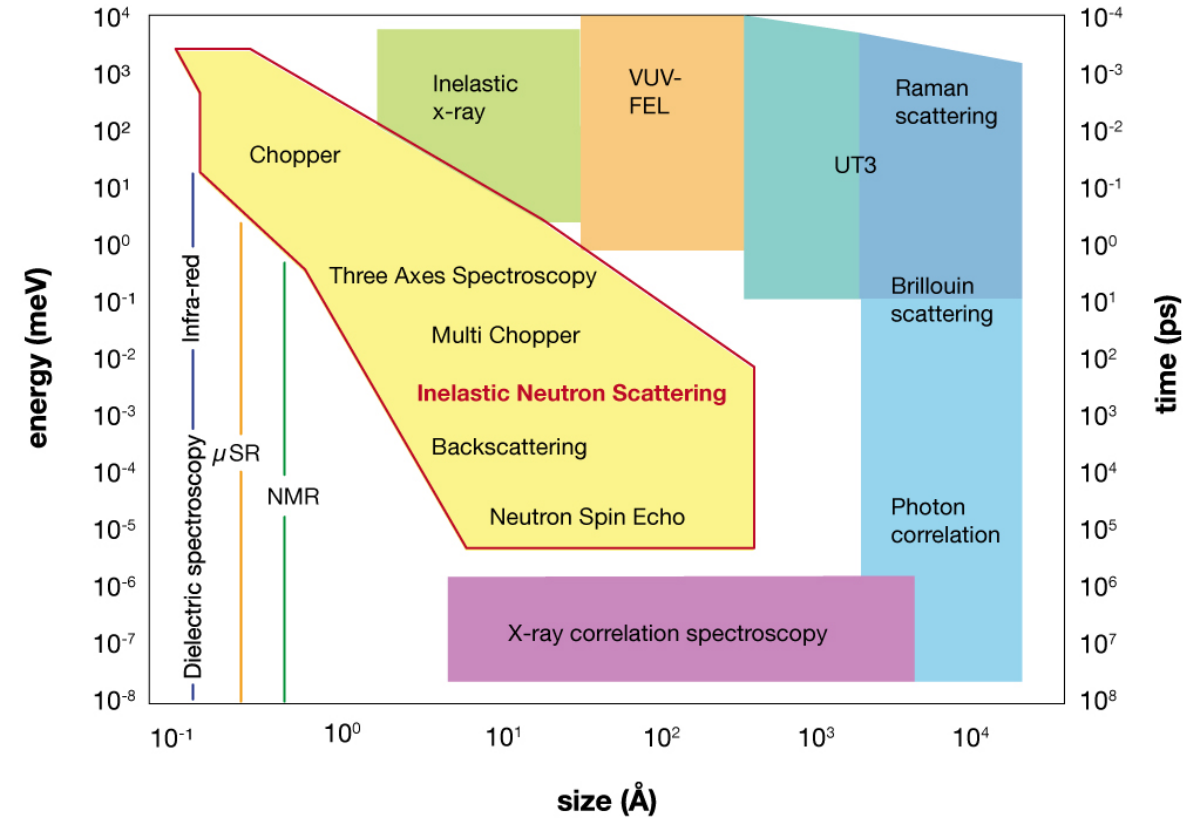
Take home message

- **Coherent, elastic scattering** shows where atoms are (**Bragg's law**)
- **Incoherent, elastic** scattering contributes to the **background** independent of angle
- **Coherent, inelastic** scattering describes the **collective movement of atoms**
- Incoherent, inelastic scattering describes diffusion (the self-correlation function of atoms)

Physics explored with neutrons



ILL yellow book

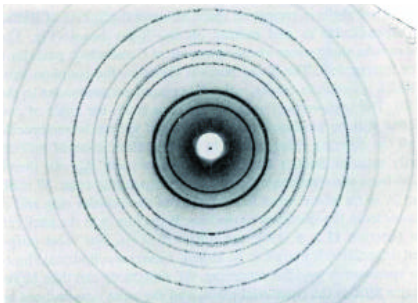
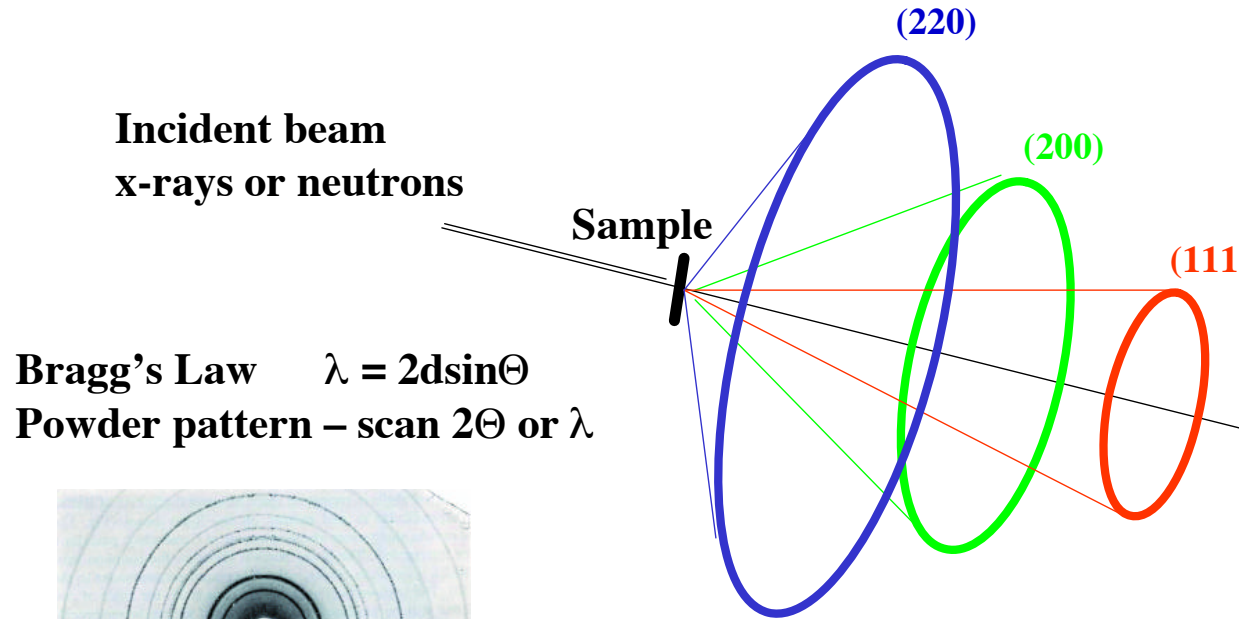


MLZ blue book

Instruments

- Elastic scattering (Diffractometer)
 - Diffractometer (Powder and single crystal)
 - Small angle scattering (SANS)
 - Reflectometer
- Inelastic scattering (spectrometer, energy transfer meV region)
 - Three axis spectrometer
 - TOF spectrometer
- Quasielastic scattering (Energy transfer in the meV region)
 - Backscattering spectrometer
 - Spin-echo spectrometer
- Imaging instruments (direct observation), nuclear and fundamental physics, Positron source, medical applications and irradiation facility

Powder Diffraction



A typical powder diffraction pattern

Pulverdiffraktometer SPODI am FRM II



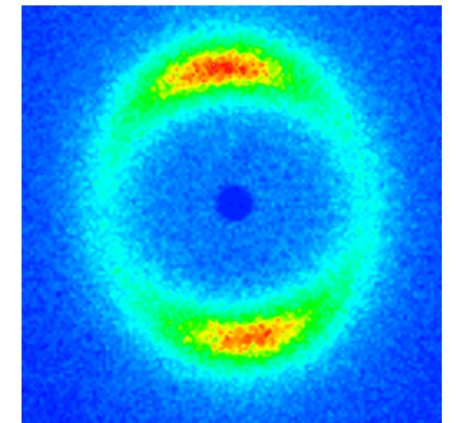
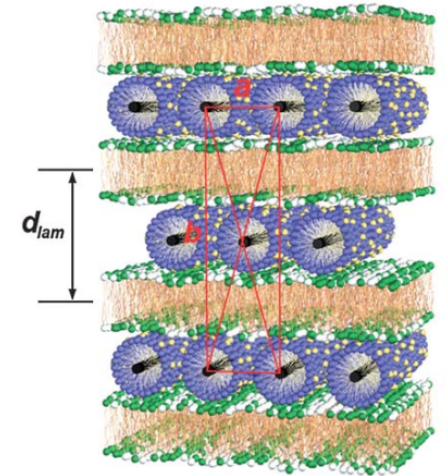
SANS - Resumé

SANS: Diffractometer specialized for small scattering angles

Large correlations in real space \longleftrightarrow Low Q small scattering angles
20 to 40000 Å $\sim 1 \text{ \AA}^{-1}$ to $\sim 10^{-4} \text{ \AA}^{-1}$

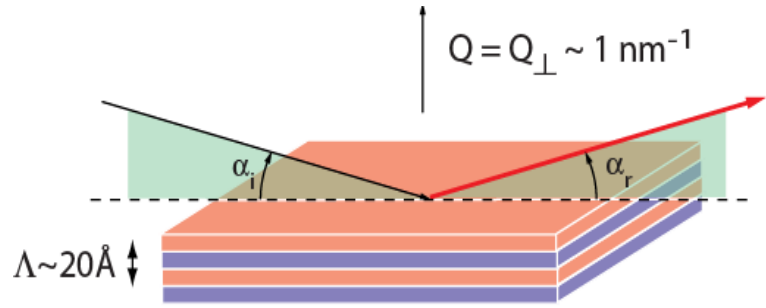
SANS tells you the statistical average of:

- **Shape** of scattering object
- **Size** (distribution) of scattering objects
- Surface of scattering objects
- Scattering length density (distribution)
- Arrangement (Superstructure?)

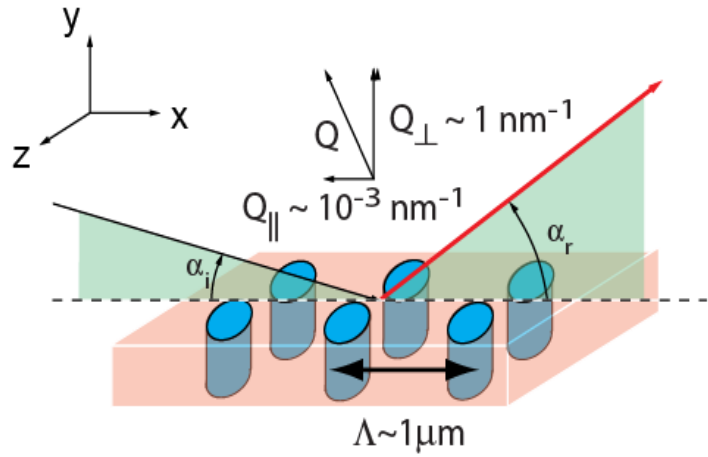
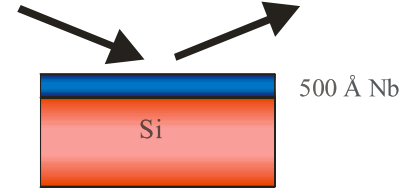
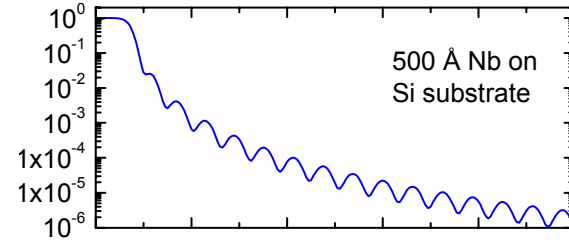


SANS 1 at FRM II

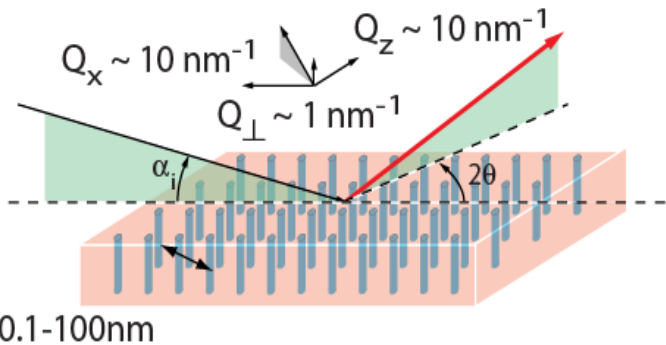
Reflectometer



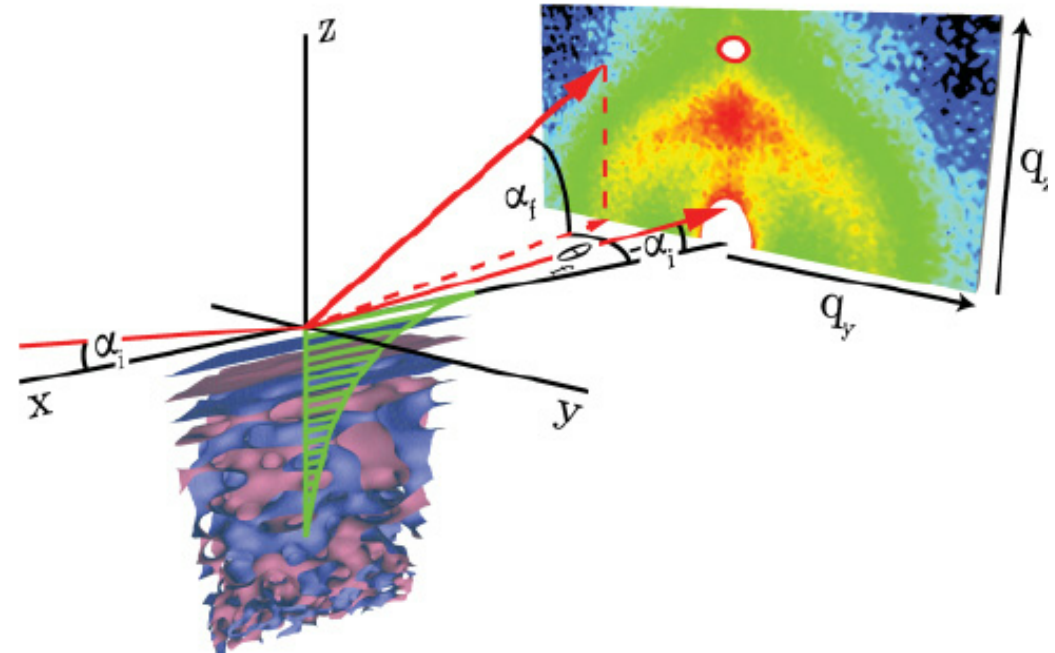
Specular reflectometry
 Depth profiles
 (nuclear and/or magnetic)



Off-specular (diffuse) scattering
 In-plane correlated roughness
 Magnetic stripes
 Phase separation (polymers)



Glancing incidence diffraction
 Ordering in liquid crystals
 Atomic structures near surfaces
 Interactions among nanodots

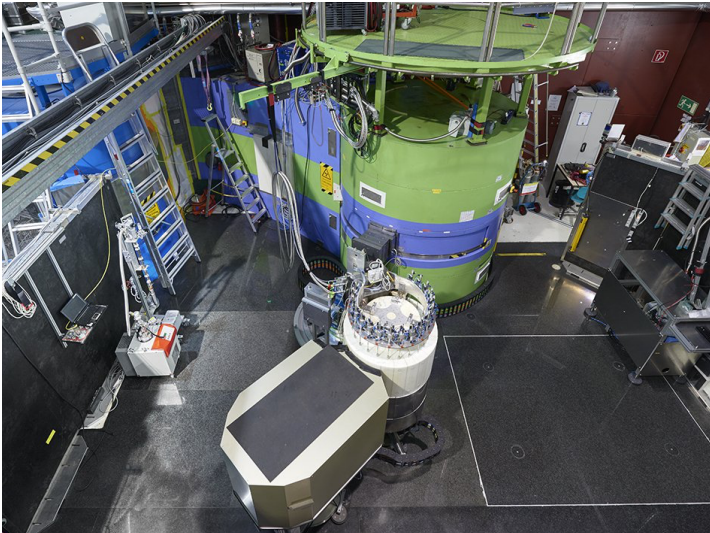


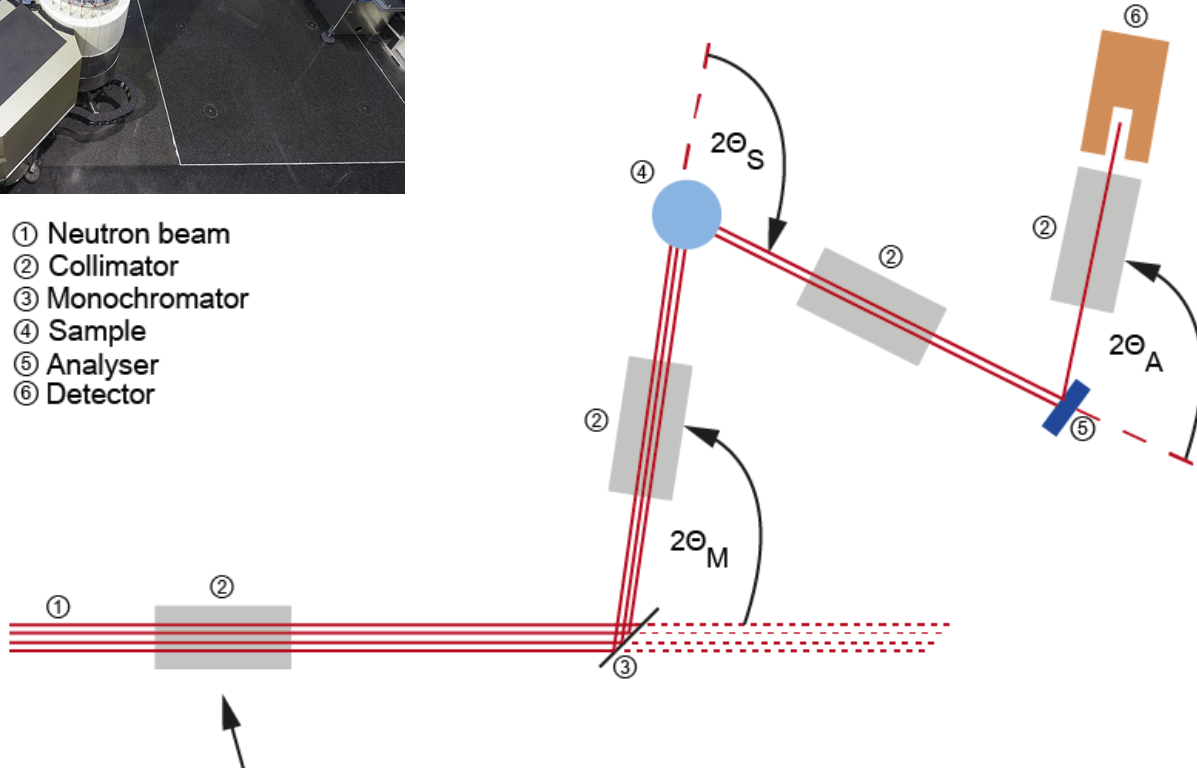
Viewgraph from M. R. Fitzsimmons

Three axis spectrometer (TAS)

PUMA@FRMII

- ▶ cold source: $\lambda \approx 5 \text{ \AA}$
- ▶ monochromator: selects initial energy E_i
- ▶ sample environment: cryostat and magnets
- ▶ analyzer: selects final energy E_f
- ▶ scattering angle: selects momentum transfer $\mathbf{Q} = \mathbf{G} + \mathbf{q}$
- ▶ maps out the dispersion relation $\hbar\omega(\mathbf{q}) = E_i - E_f$

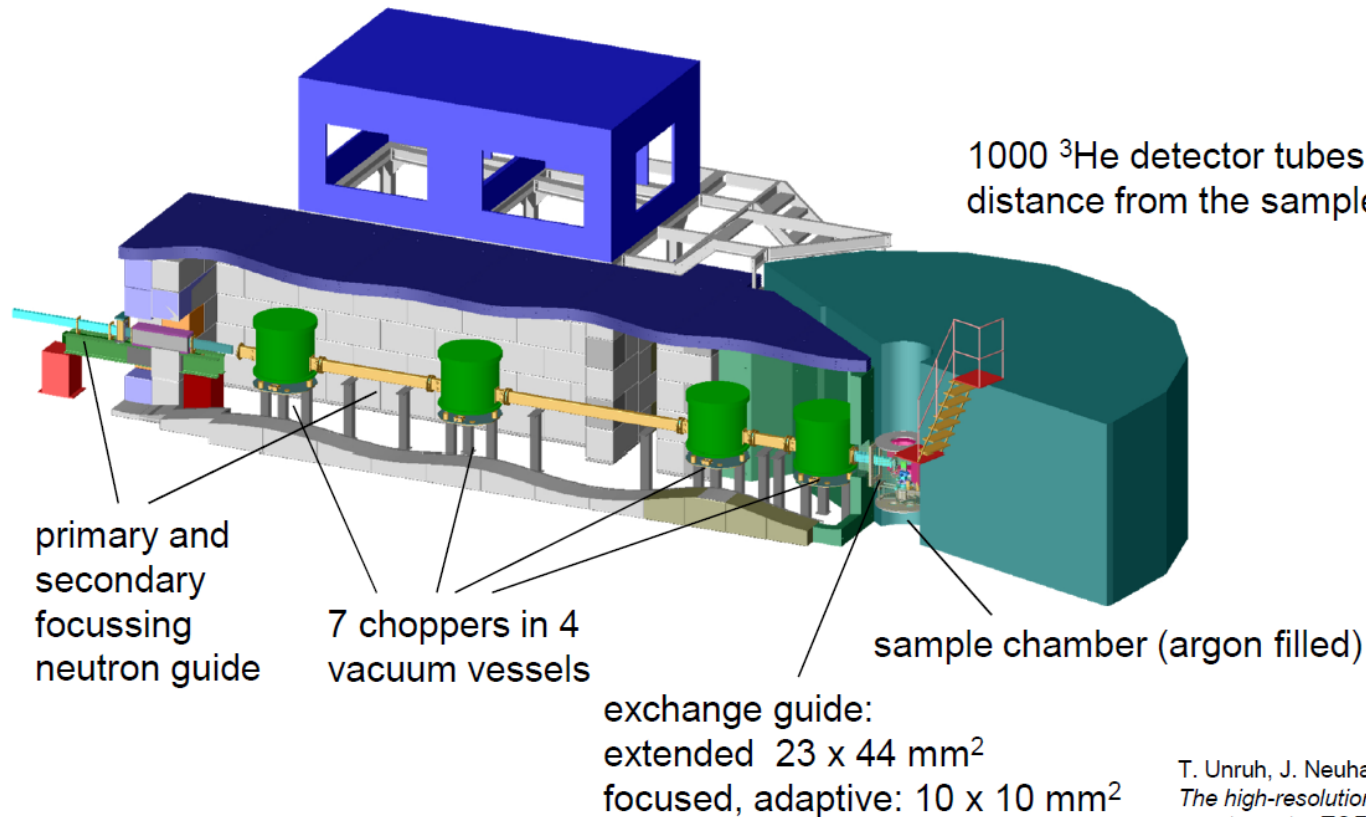
- 
- ① Neutron beam
 - ② Collimator
 - ③ Monochromator
 - ④ Sample
 - ⑤ Analyser
 - ⑥ Detector



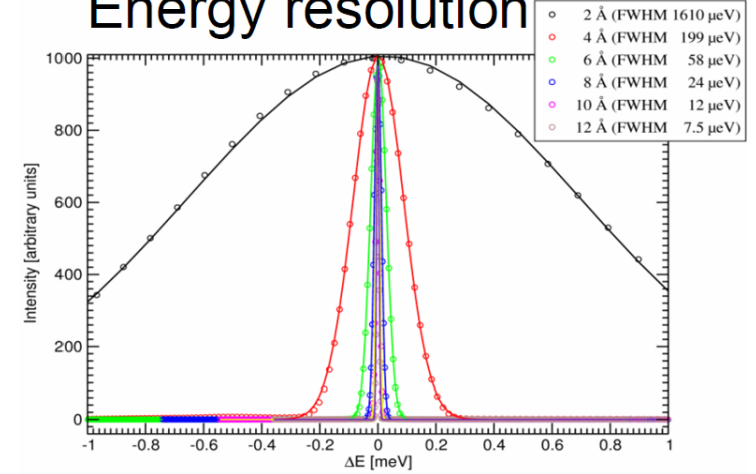
- ▶ here: keep E_i and \mathbf{q} fix and scan $\hbar\omega$ for several \mathbf{q} 's and temperatures

Time of flight spectrometer

Instrument Layout

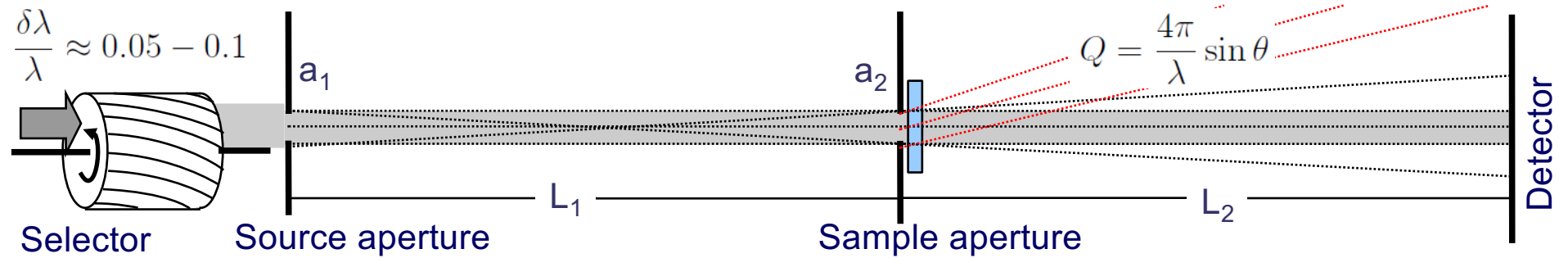


Energy resolution



T. Unruh, J. Neuhaus, W. Petry
The high-resolution time-of-flight spectrometer TOFTOF, Nucl. Instr. Methods A 580 (2007) 1414

SANS – Resolution



Angular resolution
Monochromaticity
Detector resolution
Gravity

Treat as Gaussian distributions: $\left\langle \frac{\delta Q^2}{Q^2} \right\rangle = \left\langle \frac{\delta \lambda^2}{\lambda^2} \right\rangle + \left\langle \frac{\cos^2 \theta \delta \theta^2}{\sin^2 \theta} \right\rangle$

$\left\langle \frac{\delta Q^2}{Q^2} \right\rangle = 0.0025 + \left\langle \frac{\delta \theta^2}{\theta^2} \right\rangle \Rightarrow$ Angular resolution: $\delta \theta \approx \sqrt{\frac{5}{12} \frac{a}{L}}$

What is the largest object SANS can detect (limit small Q)?

For large scattering angles (large Q) wavelength resolution dominates.

For $a_1 = a_2 = a$ and $L_1 = L_2 = L$

$\Rightarrow \delta Q \approx \frac{\delta \theta}{\theta_{min}} Q_{min} \approx \delta \theta \frac{4\pi}{\lambda} \approx \frac{2\pi a}{\lambda L}$

Largest object: $\frac{2\pi}{\delta Q} = \frac{\lambda L}{a}$

On D11, ILL: $L=40\text{m}$, $\lambda=15\text{\AA}$ $\Rightarrow D \approx 5\mu\text{m}$

Several atoms: Superposition of scattering waves

- sum over all atoms N: $\psi_{scatter} = \sum_{i,j=0\dots N} e^{-i\vec{k}_0 \cdot \vec{R}_i} \left[\frac{-b_j}{|\vec{r} - \vec{R}_j|} e^{-i\vec{k}_f \cdot (\vec{r} - \vec{R}_j)} \right]$
- simplify using: $\vec{Q} = \vec{k}_f - \vec{k}_0$, the scattering vector and
- $r \gg R_j$
- gives: $\frac{d\sigma}{d\Omega} = \sum_{i,j} b_i b_j e^{-i\vec{Q} \cdot (\vec{R}_i - \vec{R}_j)}$
- using

